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Claus Weitkamp

# Lidar

Range-Resolved Optical  
Remote Sensing of the  
Atmosphere



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Claus Weitkamp  
Editor

# **Lidar**

## **Range-Resolved Optical Remote Sensing of the Atmosphere**

Foreword by Herbert Walther

With 162 Illustrations

 Springer

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## Foreword

Soon after the laser was invented it became obvious that this new instrument, providing visible and infrared radiation with high intensity density and small divergence, would be a great tool for remote monitoring of atmospheric properties by radar-like methods. The lidar method (li for light instead of ra for radio) is thus practically as old as the laser itself. In the beginning, measurements using light scattering of aerosols and dust particles were at the focus of attention, e.g., for investigating visibility or cloud heights. As lasers became more intense, and systems for detecting backscattered light more sensitive, Rayleigh scattering was also studied to allow parameters such as variation of the total air pressure or clear air turbulence to be investigated. Furthermore, Raman scattering allowed limited selective detection of gas constituents of the atmosphere. Increasing sophistication of laser systems made it possible to study trace constituents of the atmosphere; this was the case as soon as frequency-tunable laser systems, either line-tunable or continuously tunable, became available. This made selective absorption and fluorescence accessible for detecting trace constituents with a sensitivity sufficiently high to be useful.

Improved methods such as the differential absorption method were also invented and used to monitor trace constituents over large distances. In this way, the stratospheric ozone concentration, for example, could be monitored with good accuracy and also checked by comparison with the results of other methods applied simultaneously. Today lidar ozone measurements are being routinely applied by many laboratories in the world.

Not only were laser systems improved in respect to sophistication, they also became more reliable and more efficient, so that finally, long-term routine use was possible and mobile systems, especially advantageous for pollution monitoring, came into use. Systems were also applied in airplanes and satellites, thus tremendously increasing the range of application of lidar methods.

Improvement of laser technology was vital in the lidar field. At first, researchers were mainly concerned with keeping the laser of their lidar system working; finally, they were able to concentrate on optimizing the measurement procedure itself and spend the time evaluating their data.

Lidar thus went through many ups and downs. Finally, it can be stated that the technology reached the stage where methods became reliable. With a few exceptions, e.g., the white light femtosecond lidar system, the techniques described in the present book are in principle already mentioned in previous reviews of the field. However, the experience gained in the last few years with the different methods has been hugely extended by progress in laser techniques, so that finally the enormous potential of lidar was recognized.

I am sure that this book will help newcomers to the field to obtain the information necessary for learning about the potential of lidar methods and applying the techniques for making useful measurements. Researchers working in the lidar field will profit from the practice of experienced colleagues who contributed to the book.

Herbert Walther  
University of Munich and  
Max Planck Institute of Quantum Optics  
Garching, August 2004

## Preface

This is a book on lidar, an active range-resolving optical remote measurement technique. Lidar allows us to measure, not just “sense,” virtually every property of the atmosphere. Lidar uses as a probe optical radiation, or light, even if this radiation is not always within the limits of the visible part of the spectrum. Lidar, or laser radar, as it is sometimes called, borrows its name from analogy with the better-known radar. There are indeed certain similarities, and yet the full name of which lidar is the acronym, “light detection and ranging,” is more misleading than enlightening.

In the early days lidar was considered a “poor man’s radar,” as one author put it. This is no longer the case. Today’s lidar systems have reached a high degree of maturity and sophistication. They allow scientists to measure a rich variety of atmospheric parameters on a routine basis. Lidars range in size from a shoebox to a 40-foot container. Compared to radar, lidar offers much better sensitivity to aerosols, much better spatial resolution (lateral resolution on the order of one meter at a distance of ten kilometers with no side lobes), and comparable depth and time resolution. Lidars can measure gases which radar cannot. Lidar opens perspectives for the remote, noncontact, range-resolved measurement of the element composition of aerosols and remote chemical analysis of distant targets. As a whole, lidar provides more and different information on the state of the atmosphere. Lidars and radars thus complement each other in an ideal synergism.

This book covers, for each of the major variants of the lidar technique, the underlying physics—how it works, its mathematics—what the relevant equations look like, the basic layout of an instrument, and examples of atmospheric properties that can be determined and atmospheric



processes that can be investigated. Range-finding, bathymetry, active imaging of a remote object (“lidar”), and non-range-resolving remote fluorescence measurements are not described. Also, this is not a book on meteorology, atmospheric chemistry, atmospheric physics, or pollution abatement strategies. Because the book is not a monograph, the relative importance attached by the authors to the different aspects varies: Some chapters are more theoretical, others are more descriptive in nature. Some authors stress the historical development, others devote most of the available space to applications. The style and the use of mathematical symbols are not strictly consistent from one chapter to another, although some uniformity in terminology has been achieved.

The first two chapters after the introduction each review a phenomenon of particular importance to lidar, namely, depolarization and multiple scattering. The following chapters are devoted either to a lidar variant that is based on one type of interaction such as elastic scattering, absorption, or Raman scattering, or to the measurement of one particular atmospheric property (visibility, moisture). Some of the chapters contain original material that cannot be found in books or archival journals, but is presented here for the first time. I should like to depict this with a few examples.

For example, in the chapter on polarization (Chapter 2), the contents of Figures 2.3, 2.4, and 2.5, the discussion about Saharan dust in Florida, and the description of the effect of clouds are original material, although some of it has been used in articles that are about to appear or have appeared recently. Chapter 3, on multiple scattering, is another example. It is the first known attempt to assemble in logical sequence, under a single cover, our current knowledge on modeling, correcting for, and exploiting multiple scattering in lidar. The material discussed was hitherto dispersed in numerous publications, and some of it was not very well known and consequently little used; an example is the effective-medium theorem of Section 3.3.3, which not only constitutes an important theoretical result by itself but allows considerable simplifications in practical applications. Chapter 4 is a review of the formidable efforts made to extract aerosol microphysical data from lidar signal profiles. Whereas the (forward) calculation of an expected lidar signal from a given profile of aerosol properties is straightforward, the inverse problem, ill posed in mathematical terms, is nevertheless successfully tackled. The description of these efforts in a balanced, comprehensible representation is not found elsewhere in the literature. The same applies to the use of Raman

scattering and high-spectral-resolution lidar (Chapter 5) for the determination of aerosol properties. Visual range is a quantity that is clearly defined, yet in different ways for different applications. These definitions are presented here in a clear-cut, yet concise, approach. The visibility chapter (Chapter 6) is thus more a description of the actual state of the art, stressing modern developments such as the intelligent automobile taillight, heliport (miniaturized) visibility lidar for quicker helicopter rescue logistics, and a soon-to-come web-based, real-time, global-coverage cloud-bottom-height and visibility-profile information network. Chapter 7, on differential-absorption lidar (DIAL), also strongly exhibits a review character, grouping techniques according to wavelength and, within one wavelength range, history; this representation was chosen because techniques differ greatly with wavelength range. Chapter 8, also on DIAL, deals with two gases only but two that are particularly hard to measure. It therefore contains the in-depth treatment of effects important for water vapor and oxygen DIAL and present, although of lesser importance, in DIAL of other gases as well. In Chapter 9, the equations that describe Raman scattering are reformulated with special emphasis on the needs of the lidar scientist. In doing so, the author also implemented the transition from the cgs to the SI system of units. Again, this work has not been published elsewhere. The same applies to the comprehensive theory and performance simulations of Chapter 10 on temperature measurements with lidar in general and rotational Raman lidar in particular; the complete optimization calculations are made available here for the first time. Chapters 11 and 12, on fluorescence lidar and Doppler wind lidar, are kept relatively short, describing the mechanisms and general features of the techniques, but still giving a very few examples of applications: composition, wind, temperature of the upper atmosphere, gravity waves, and meteor science in the case of fluorescence lidar; clear-air turbulence, wind turbines, aviation safety, weather forecasting, and other synergisms for Doppler wind lidar. Chapter 13 is a survey of airborne and spaceborne lidars and their applications, past, present, and future. Again, we know of no other similarly comprehensive review of the subject. Chapter 14, finally, presents two examples of recent lidar developments. For the first, broadband-emission lidar with narrow-band determination of absorption, the complete theory is shown here for the first time. The second, terawatt-femtosecond white-light lidar, opens a path to the remote determination of hitherto inaccessible properties and offers exciting perspectives for atmospheric research through qualitatively new applications of remote measurement technology.

The idea of the book was first discussed in a round-table meeting on the occasion of the Twenty-First International Laser Radar Conference in Quebec City, Quebec, Canada, on 12 July 2002. The time it took to publish the book may appear quite long. In view of the workload imposed today on researchers in science and technology it is amazing that it did not take longer and, more important, that all relevant variants of lidar could be covered.

The authors come from five different nations. They are all world-renowned lidar experts. However, the origin of the authors of this book has nothing to do with the effort and ingenuity devoted by different countries to work in the field. Otherwise, such important lidar nations as Britain, China, Italy, the Netherlands, Russia, Sweden, and Switzerland would not be unrepresented, to name just a few. The same is true for the citations in the different chapters: space did not allow reference to all important contributions, so even some of the most significant, pioneering work had to be left out.

Support by GKSS during the preparation of the manuscript is gratefully acknowledged. Still, this book would not have come into existence without many fruitful discussions with colleagues at Geesthacht and elsewhere. Most authors took a greater than usual interest in the progress of the book, providing, in addition to their manuscripts, many useful suggestions, help, confidence, and moral support. The same is true of Hans Koelsch, volume editor of the publishing company. I thank them all.

Claus Weitkamp  
Geesthacht, Germany  
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# 1

## Introduction to Lidar

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### 1.1 Lidar and the Atmosphere

Atmospheric research nowadays is hard to conceive without the use of remote-sensing techniques. Light detection and ranging (lidar) is, along with radiowave detection and ranging (radar), one of the backbones of the research field that deals with the profiling of the atmosphere. High spatial and temporal resolution of the measurements, the possibility of observing the atmosphere at ambient conditions, and the potential of covering the height range from the ground to more than 100 km altitude make up the attractiveness of lidar instruments. The variety of interaction processes of the emitted radiation with the atmospheric constituents that can be used in lidar allow the determination of the basic atmospheric variables of state, i.e., temperature, pressure, humidity, and wind, as well as the measurement of trace gases, aerosols, and clouds.

Lidar has largely contributed to our knowledge of the Earth's atmosphere during the past decades. It is particularly useful for the investigation of highly variable atmospheric parameters. Lidar has the potential for the observation of processes on scales that extend from a few cubic meters and a few seconds to global, multi-year coverage. Lidar has been used to investigate turbulent processes and the diurnal cycle of the planetary boundary layer, including the measurement of water-vapor and ozone fluxes. Meteorological phenomena such as frontal passages, hurricanes, and mountain lee waves were studied. Lidar helps monitor emission rates and concentration levels of trace gases. The stratospheric ozone depletion is documented globally with lidar. The role of polar stratospheric clouds is investigated and the classification of

polar stratospheric clouds is based on their scattering properties as seen with lidar. Lidar is used to distinguish water droplets from ice crystals in clouds. Lidar contributes to our knowledge of the climatic effects of aerosols. The stratospheric perturbation after major volcanic eruptions has been studied and the intercontinental transport of air pollution, desert dust, and forest-fire smoke has been detected. In the mesosphere, lidar has proven the existence of layers of metallic atoms and ions and of gravity waves therein. Lidar instruments can operate from the ground or from aircraft, one system has been flown on the Space Shuttle, and in the near future satellite-based lidar instruments will carry out global observations of atmospheric constituents from space. These and many more aspects of lidar are presented in this book giving an overview on the state of the art of the basic lidar techniques used in the investigation of the Earth's atmosphere.

## 1.2 Lidar History

The introduction of the lidar principle dates back to pre-laser times. In the 1930s first attempts were made to measure air density profiles in the upper atmosphere by determining the scattering intensity from searchlight beams [1–4]. Height information was obtained by scanning the receiver field of view of a distant telescope along the continuous light beam [5]. In 1938, pulses of light were used for the first time to measure cloud base heights [6]. The generation of light pulses by electric sparks and flashlamps allowed the replacement of the bistatic configuration by a monostatic setup, i.e., a setup in which transmitter and receiver are collocated and the height information is actively deduced from a measurement of the round-trip time between pulse emission and signal detection. The acronym lidar for this kind of measurement technique was first introduced by Middleton and Spilhaus in 1953 [7].

The rapid development of modern lidar technology started with the invention of the laser in 1960 [8] and the giant-pulse or Q-switched laser in 1962 [9]. Fiocco and Smullin published atmospheric observations with a ruby laser in 1963 [10]. About a decade later all basic lidar techniques had been suggested and demonstrated. Consequently, the first textbook on lidar edited by E.D. Hinkley [11] appeared in 1976. Ever since, success in lidar development was strongly connected with progress in optical and electronic technology, in particular

laser technology. Lidar researchers have always been involved in laser development. Many instruments use lasers specifically designed for lidar to meet the high requirements of certain lidar techniques on laser power, wavelengths, pulse width, beam shape, and spectral purity often not fulfilled by commercial products. In addition to lasers, optical filters with high transmissivity, narrow bandwidth, steep spectral slopes and/or high out-of-band suppression, efficient detectors for broad wavelength regions, data-acquisition systems with a dynamic range of several orders of magnitude, and computers that can process large amounts of data with high repetition rate belong to the devices needed for advanced lidar systems. Lidar has therefore always been both a source and a beneficiary of technological innovation.

### 1.3 Lidar Setup

The basic setup of a lidar system is shown in Fig. 1.1. In principle, a lidar consists of a transmitter and a receiver. Short light pulses with lengths of a few to several hundred nanoseconds and specific spectral properties are generated by the laser. Many systems apply a beam expander within the transmitter unit to reduce the divergence of the light beam before it is sent out into the atmosphere. At the receiver end, a telescope collects the photons backscattered from the atmosphere. It is usually followed by an optical analyzing system which, depending on the application, selects specific wavelengths or polarization states out of the collected light. The selected radiation is directed onto a detector,

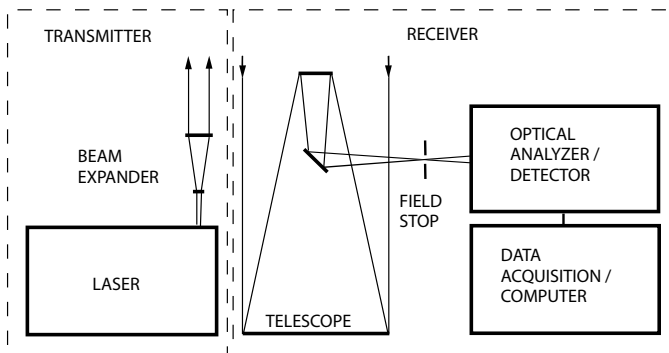


**Fig. 1.1.** Principle setup of a lidar system.

where the received optical signal is converted into an electrical signal. The intensity of this signal in its dependence on the time elapsed after the transmission of the laser pulse is determined electronically and stored in a computer.

Wavelengths used in lidar depend on the application and extend from about 250 nm to 11  $\mu\text{m}$ . While ruby, nitrogen, copper-vapor, and  $\text{CO}_2$  lasers were mainly used in the early years, high-power excimer and Nd:YAG lasers have been spreading out in the field since the 1980s. Excimer lasers produce ultraviolet radiation, whereas the Nd:YAG crystal emits in the infrared spectral region at a wavelength of 1064 nm. Frequency doubling and tripling with nonlinear crystals is widely used to convert the primary Nd:YAG radiation to 532 and 355 nm. Quadrupling to 266 nm is also utilized. Both laser types serve not only as direct lidar emitters, but also to pump secondary laser sources. The radiation can be shifted to longer wavelengths by stimulated Raman scattering in gases such as hydrogen and deuterium. This technique is preferably applied in the ultraviolet for ozone differential-absorption lidar and solar-blind Raman lidar. Dye lasers pumped either with excimer or Nd:YAG lasers have been used for a long time to produce the specific wavelengths needed for differential-absorption and resonance-fluorescence lidar. Nowadays, they are more and more being replaced by tunable, solid-state lasers based, e.g., on titanium:sapphire or alexandrite crystals and by optical parametric oscillators. The doping of crystalline lattices, e.g., yttrium aluminium garnet (YAG), yttrium lithium fluoride (YLF), lutetium aluminium garnet (LuAG), or of glasses with active ingredients such as Nd, Ho, Tm, Cr, Er, or Yb, creates a wide range of infrared wavelengths, some of which are particularly well suited for Doppler lidar. Presently, new laser types such as slab, microchip, waveguide, and solid-state Raman lasers are under investigation for their possible use in lidar.

Although laser beams are already highly collimated, their divergence is often further reduced by beam expansion to values of the order of 100  $\mu\text{rad}$ . Then, the field of view of the receiver telescope can be chosen as low as a few hundred  $\mu\text{rad}$ . This has several benefits. First of all, background light from the atmosphere is efficiently reduced. Second, fewer photons that underwent multiple scattering in the atmosphere are detected. Finally, a small field of view is necessary for lidar methods based on signal detection with high spectral resolution because of the small acceptance angles of the wavelength-selective optical devices. Depending on the purpose of the lidar, the diameter



of the primary telescope optics ranges from 0.1 to a few meters. The majority of lidars use mirror telescopes. Lenses can only be used for small-aperture receivers. The field of view is determined by a field stop in the focal plane of the receiver optics. Lidar systems for investigations of the higher atmosphere utilize a chopper at this position. The chopper opens the field stop only when light from the region of interest arrives and thus blocks the strong backscatter signal from the lower atmosphere to avoid an overload of the detectors.

The geometric arrangement of the emitter and receiver optics determines the degree of signal compression at distances close to the lidar. At short distances the laser beam cannot completely be imaged onto the detector. Thus, only a part of the actual lidar return signal is measured. This part varies with distance and depends on laser beam diameter, shape, and divergence, the telescope's imaging properties (focal-length-to-diameter ratio), the receiver field of view, and the location of emitter and receiver optical axes relative to each other. In coaxial systems the laser beam is emitted along the optical axis of the receiver telescope. In biaxial systems the optical axes are spatially separated by at least one radius of the telescope mirror, and the laser beam enters the telescope field of view from the side. The function resulting from the combination of all geometric effects is called the laser-beam receiver-field-of-view overlap function. Its value is zero at the lidar and becomes unity when the laser beam is completely imaged onto the detector through the field stop. For large telescopes the overlap function can affect the lidar return signal up to distances of several kilometers.

Optical analysis of the backscattered light is usually done before the detection. In the simplest case, an interference filter is placed in front of the detector. The filter transmits light in a certain passband around the wavelength of interest and suppresses light outside the transmission band, e.g., background radiation. Other applications require more sophisticated solutions for the spectral analysis. Polarizers, grating spectrometers, interferometers, and atomic-vapor filters belong to the elements applied. Some examples are explained in more detail below.

Signal detection is realized with photomultiplier tubes (PMTs) or photodiodes. With PMTs and avalanche photodiodes (APDs) operated in the Geiger mode photons can be counted individually. The photon-counting technique is very sensitive and is used when the backscatter signal is weak, e.g., when it results from a weak scattering process or when the investigated region is far away from the instrument. The number

of photon counts per time interval after emission of the laser pulse is stored. The resolved time interval  $\Delta t$  corresponds to an atmospheric range bin  $\Delta R = c\Delta t/2$  given by the speed of light  $c$  and the factor  $1/2$  because the light has to travel forth and back. Storing signals with a time resolution of 100 ns thus results in an atmospheric range resolution of 15 m, provided the laser pulse is shorter than  $\Delta R$ . For strong backscatter signals, analog recording is the method of choice, i.e., the average current produced by the photo pulses is measured, followed by analog-to-digital (AD) signal conversion and digital signal processing. In this case, the AD conversion rate determines the achievable range resolution of the system. Laser pulse repetition rates range from a few up to several thousand shots per second. Because the corresponding high time resolution is not meaningful, lidar signals are normally averaged over time intervals of a few seconds to minutes to reduce the amount of data that must be stored. A few applications require single-shot processing before averaging and need fast computer processing.

## 1.4 Lidar Equation

In the simplest form, the detected lidar signal can be written as

$$P(R) = K G(R) \beta(R) T(R), \quad (1.1)$$

i.e., the power  $P$  received from a distance  $R$  is made up of four factors. The first factor,  $K$ , summarizes the performance of the lidar system, the second,  $G(R)$ , describes the range-dependent measurement geometry. These two factors are completely determined by the lidar setup and can thus be controlled by the experimentalist. The information on the atmosphere, and thus all the measurable quantities, are contained in the last two factors of Eq. (1.1). The term  $\beta(R)$  is the backscatter coefficient at distance  $R$ . It stands for the ability of the atmosphere to scatter light back into the direction from which it comes.  $T(R)$  is the transmission term and describes how much light gets lost on the way from the lidar to distance  $R$  and back. Both  $\beta(R)$  and  $T(R)$  are the subjects of investigation and in principle unknown to the experimentalist.

Going into more detail, we can write the system factor as

$$K = P_0 \frac{c\tau}{2} A\eta. \quad (1.2)$$

$P_0$  is the average power of a single laser pulse, and  $\tau$  is the temporal pulse length. Hence  $E_0 = P_0\tau$  is the pulse energy, and  $c\tau$  is the length of the volume illuminated by the laser pulse at a fixed time. The factor  $1/2$  appears because of an apparent “folding” of the laser pulse through the backscatter process as illustrated in Fig. 1.2. When the lidar signal is detected at an instant time  $t$  after the leading edge of the pulse was emitted, backscattered light from the leading edge of the pulse comes from the distance  $R_1 = ct/2$ . At the same time, light produced by the trailing edge arrives from distance  $R_2 = c(t - \tau)/2$ . Thus  $\Delta R = R_1 - R_2 = c\tau/2$  is the length of the volume from which backscattered light is received at an instant time and is called the “effective (spatial) pulse length.”  $A$  is the area of the primary receiver optics responsible for the collection of backscattered light, and  $\eta$  is the overall system efficiency. It includes the optical efficiency of all elements the transmitted and received light has to pass and the detection efficiency. The telescope area  $A$  and the laser energy  $E_0$ , or, rather, the average laser power  $\bar{P} = E_0 f_{\text{rep}}$ , with the pulse repetition frequency  $f_{\text{rep}}$ , are primary design parameters of a lidar system. The experimentalist will also try to optimize the overall system efficiency  $\eta$  to obtain the best possible lidar signal.

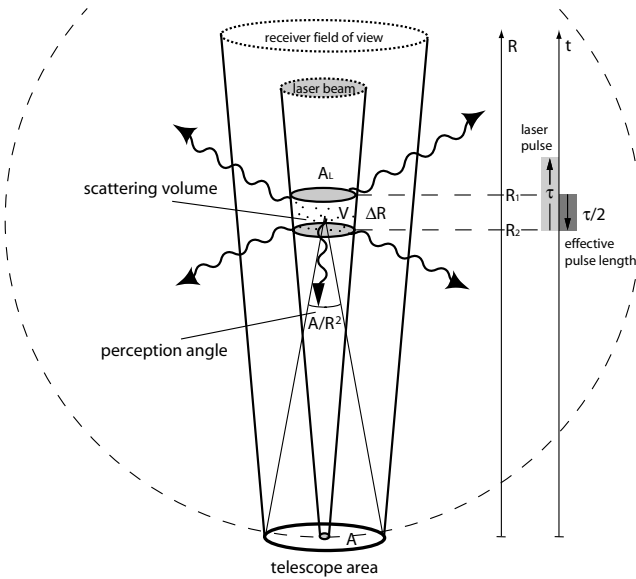


**Fig. 1.2.** Illustration of the lidar geometry.

The geometric factor

$$G(R) = \frac{O(R)}{R^2} \quad (1.3)$$

includes the laser-beam receiver-field-of-view overlap function  $O(R)$  described before and the term  $R^{-2}$ . The quadratic decrease of the signal intensity with distance is due to the fact that the receiver telescope area makes up a part of a sphere's surface with radius  $R$  that encloses the scattering volume (see Fig. 1.2). If we imagine an isotropic scatterer at distance  $R$ , the telescope area  $A$  will collect the fraction

$$\frac{I_c}{I_s} = \frac{A}{4\pi R^2} \quad (1.4)$$

of the overall intensity  $I_s$  scattered into the solid angle  $4\pi$ . In other words, the solid angle  $A/R^2$  is the perception angle of the lidar for light scattered at distance  $R$ . The factor  $4\pi$  does not appear explicitly in the lidar equation because it cancels out by the definition of the backscatter coefficient  $\beta$  as we will see below. It is primarily the  $R^{-2}$  dependence that is responsible for the large dynamic range of the lidar signal. If we start detecting a signal with  $O(R) = 1$  at a distance of 10 m, the signal will be 6 orders of magnitude lower at 10 km distance just because of the geometry effect. To what extent lidar is a *range-resolving and remote* measurement technique depends on our ability to compensate for this effect. Geometrical signal compression at short distances is one possibility as can be seen from Fig. 1.3 in which an arbitrary, but realistic overlap function is shown, multiplied with the function  $R^{-2}$ . The strong signal in the near field is suppressed by several orders of magnitude. On a few occasions the atmosphere will help in compressing the signal by an increase of the backscattering at larger distances as we will see below. In most cases, however, the atmosphere causes an additional decrease of the signal with range.

The backscatter coefficient  $\beta(R, \lambda)$  is the primary atmospheric parameter that determines the strength of the lidar signal. It describes how much light is scattered into the backward direction, i.e., towards the lidar receiver. The backscatter coefficient is the specific value of the scattering coefficient for the scattering angle  $\theta = 180^\circ$ . Let  $N_j$  be the concentration of scattering particles of kind  $j$  in the volume illuminated by the laser pulse, and  $d\sigma_{j,\text{sca}}(\pi, \lambda)/d\Omega$  the particles' differential scattering cross section for the backward direction at wavelength  $\lambda$ . The

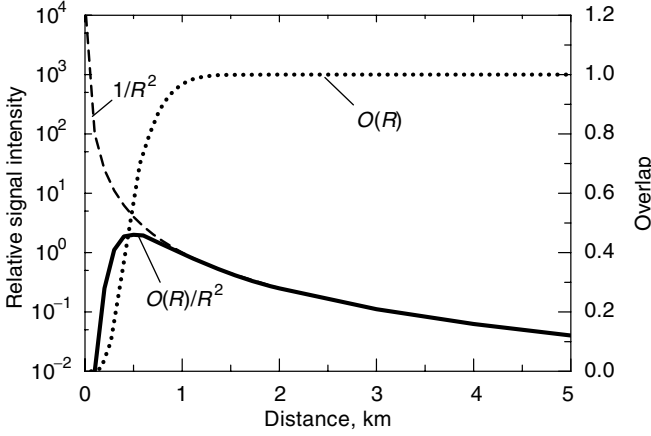


**Fig. 1.3.** Influence of the overlap function on the signal dynamics.

backscatter coefficient can then be written as

$$\beta(R, \lambda) = \sum_j N_j(R) \frac{d\sigma_{j,\text{sca}}}{d\Omega}(\pi, \lambda), \quad (1.5)$$

with summing over all kinds of scatterers. Since the number concentration is given in units of  $\text{m}^{-3}$  and the differential scattering cross section in  $\text{m}^2\text{sr}^{-1}$ , the backscatter coefficient has the unit  $\text{m}^{-1}\text{sr}^{-1}$ .

If we return to our simplified picture of isotropic scattering and assume that there is only one type of particle in the scattering volume, the relation between the backscatter coefficient and the isotropic scattering cross section  $\sigma_{\text{sca}}$  is  $4\pi\beta = N\sigma_{\text{sca}}$ . The intensity of scattered light from the illuminated volume  $V = A_L \Delta R = A_L c\tau/2$ , with the laser-beam cross section  $A_L$ , is proportional to the area  $A_s = N\sigma_{\text{sca}}V$ , i.e., the scattering cross section of all particles in the volume  $V$ . Thus, the relative intensity of the scattered light is

$$\frac{I_s}{I_0} = \frac{A_s}{A_L} = \frac{N\sigma_{\text{sca}}c\tau}{2} = \frac{4\pi\beta c\tau}{2}. \quad (1.6)$$

With Eq. (1.4), we obtain the ratio of the collected to the emitted light intensity

$$\frac{I_c}{I_0} = \frac{A\beta c\tau}{2R^2}. \quad (1.7)$$

The right side of this equation describes that part of the lidar equation that directly refers to the scattering geometry, i.e., it contains the size and the backscatter properties of the scattering volume and the perception angle of the lidar.

In the atmosphere, the laser light is scattered by air molecules and particulate matter, i.e.,  $\beta(R, \lambda)$  can be written as

$$\beta(R, \lambda) = \beta_{\text{mol}}(R, \lambda) + \beta_{\text{aer}}(R, \lambda). \quad (1.8)$$

Molecular scattering (index mol), mainly occurring from nitrogen and oxygen molecules, primarily depends on air density and thus decreases with height, i.e., backscattering decreases with distance if the observation is made from the ground, but increases in the case of downward-looking systems on aircraft or spacecraft. Particulate scattering (index aer for aerosol particles) is highly variable in the atmosphere on all spatial and temporal scales. Particles represent a great variety of scatterers: tiny liquid and solid air-pollution particles consisting of, e.g., sulfates, soot and organic compounds, larger mineral-dust and sea-salt particles, pollen and other biogenic material, as well as comparably large hydrometeors such as cloud and rain droplets, ice crystals, hail, and graupel.

As the final part of the lidar equation, we have to consider the fraction of light that gets lost on the way from the lidar to the scattering volume and back. The transmission term  $T(R)$  can take values between 0 and 1 and is given by

$$T(R, \lambda) = \exp \left[ -2 \int_0^R \alpha(r, \lambda) dr \right]. \quad (1.9)$$

This term results from the specific form of the Lambert–Beer–Bouguer law for lidar. The integral considers the path from the lidar to distance  $R$ . The factor 2 stands for the two-way transmission path. The sum of all transmission losses is called light extinction, and  $\alpha(R, \lambda)$  is the extinction coefficient. It is defined in a similar way as the backscatter coefficient as the product of number concentration and extinction cross section  $\sigma_{j,\text{ext}}$  for each type of scatterer  $j$ ,

$$\alpha(R, \lambda) = \sum_j N_j(R) \sigma_{j,\text{ext}}(\lambda). \quad (1.10)$$

Extinction can occur because of scattering and absorption of light by molecules and particles. The extinction coefficient therefore can be

written as the sum of four components,

$$\alpha(R, \lambda) = \alpha_{\text{mol},\text{sca}}(R, \lambda) + \alpha_{\text{mol},\text{abs}}(R, \lambda) + \alpha_{\text{aer},\text{sca}}(R, \lambda) + \alpha_{\text{aer},\text{abs}}(R, \lambda), \quad (1.11)$$

where the indices sca and abs stand for scattering and absorption, respectively. Because scattering into all directions contributes to light extinction, the (integral) scattering cross section  $\sigma_{\text{sca}}$ , together with the absorption cross section  $\sigma_{\text{abs}}$ , both in  $\text{m}^2$ , make up the extinction cross section,

$$\sigma_{\text{ext}}(\lambda) = \sigma_{\text{sca}}(\lambda) + \sigma_{\text{abs}}(\lambda). \quad (1.12)$$

Consequently, the extinction coefficient has the unit  $\text{m}^{-1}$ .

As indicated in the equations above, both  $\beta$  and  $\alpha$  depend on the wavelength of the laser light. This wavelength dependence is determined by the size, the refractive index, and the shape of the scattering particles. We will discuss the consequences in conjunction with the description of the basic lidar techniques below.

Summarizing the discussion of the individual terms, we can now write the lidar equation (1.1) in a more common form as

$$P(R, \lambda) = P_0 \frac{c\tau}{2} A\eta \frac{O(R)}{R^2} \beta(R, \lambda) \exp\left[-2 \int_0^R \alpha(r, \lambda) dr\right]. \quad (1.13)$$

This equation will be used, in the one or other variation, in the following chapters as the starting point of the description of the individual lidar techniques. One should mention that the detected signal will always consist of a background contribution  $P_{\text{bg}}$  in addition to the lidar signal described above. At daytime, the background signal is dominated by direct or scattered sunlight, whereas at nighttime the moon and the stars as well as artificial light sources contribute to the background light. Detector noise is another source of undesired signal. The background must be subtracted before a lidar signal can be evaluated further. Usually, a number of data points from either the far end of the signal, where no backscattered photons are expected any more, or from the period preceding the laser pulse emission are used to calculate the mean background signal  $\bar{P}_{\text{bg}}$  and the corresponding error  $\Delta P_{\text{bg}}$  needed to compute the error of any quantity derived from the detected signals.

## 1.5 Lidar Techniques and the Contents of This Book

The contents of the book are arranged along the five basic lidar techniques which make use of specific interaction processes of the emitted radiation with the atmospheric constituents and which are:

- elastic-backscatter lidar,
- differential-absorption lidar,
- Raman lidar,
- (resonance) fluorescence lidar,
- Doppler lidar.

Each of the following chapters is self-sufficient and can in principle be understood without knowing any other chapter. It is therefore left to the reader which chapters he or she is interested to study and in which order. Chapters 2 and 3 of the book deal with polarization and multiple scattering, effects that, to a greater or lesser extent, play a role in all variants of the lidar technique. Chapters 4 to 12 describe the basic concepts, technical implications, and results that can be obtained with the most common types of lidar, following the classification given above. Chapter 13 is devoted to airborne and spaceborne applications, and chapter 14 introduces two techniques that are not yet widely applied. These two final chapters can be seen as a kind of outlook and motivation for further research on lidar techniques.

**Elastic-backscatter lidar** is the classic form of lidar and has in principle been fully described in the previous sections. In its simplest form it applies one laser emitting a single wavelength and one detector measuring the radiation elastically backscattered from the atmospheric molecules and particles. By elastic scattering we understand a process in which the wavelength of the radiation remains unchanged. This type of lidar delivers information on the presence and location of aerosol and cloud layers and is often called a Rayleigh–Mie lidar. We should, however, be careful when using this term and clarify what exactly we mean by Rayleigh and Mie scattering.

*Rayleigh scattering* can be defined as the elastic scattering from particles that are very small compared to the wavelength of the scattered radiation. In the context of lidar, Rayleigh scattering is always used as a synonym for molecular scattering. Since nitrogen and oxygen make up about 99% of the Earth's molecular atmosphere, we normally consider these two gases as the source of Rayleigh-scattered radiation.



The Rayleigh scattering intensity is proportional to  $\lambda^{-4}$  and dominates elastic-backscatter signals at short laser wavelengths. Somewhat different definitions exist concerning the spectral width of Rayleigh scattering. Temperature, pressure, and collective motion of the molecules lead to spectral broadening of the scattered radiation. The resulting intensity distribution of the elastically scattered light is called the Cabannes line. It has a spectral width of a few GHz or about  $0.1 \text{ cm}^{-1}$ . If the elastic-backscatter signal is detected with low spectral resolution, rotational Raman scattering with much higher shifts, of the order of  $100 \text{ cm}^{-1}$ , also contributes to the measured intensity. The sum of the Cabannes line and the rotational Raman bands on either side of it is therefore often referred to as Rayleigh scattering. On the other hand, Raman scattering is understood as an inelastic scattering process which involves the change of the energetic state of the molecule. From the point of view of Raman scattering theory, Rayleigh scattering is defined as the elastic scattering of radiation without a change of the vibrational-rotational quantum state of the interacting molecules and thus excludes rotational Raman scattering. We did not rule out one of the definitions in this book. Because of historic reasons and personal taste the reader will be confronted with both definitions in the literature anyhow. We tried, however, to make clear what the actual definition of the term is in the individual chapters.

Also the term *Mie scattering* is often used in a misleading way in the literature. The scattering theory developed by Gustav Mie [12] gives the analytical solution for scattering of radiation of arbitrary wavelength by a sphere of arbitrary radius and arbitrary complex refractive index. Thus, Mie scattering theory is not limited to a certain size of the scatterers, it even includes the solution for Rayleigh scattering. The term, however, is often used to describe the scattering from particles with sizes comparable to the wavelength of the radiation, or larger. The wavelength dependence of the scattered intensity is a function of particle radius relative to the wavelength and of the particles' complex refractive index. Small (Rayleigh) scatterers show the  $\lambda^{-4}$  dependence mentioned above. Scattering from very large particles does not depend on wavelength. In the region where particle radius and wavelength are of similar magnitude, the wavelength dependence of the scattering intensity varies strongly. Wavelength-dependent detection of light scattering can therefore be used to obtain information on size and other parameters of atmospheric aerosol particles in the radius range from about 50 nm to a few micrometers. The application of this technique requires

the emission of several laser wavelengths and an independent determination of the backscatter and extinction coefficients. Chapter 4 gives an overview on lidar measurements of aerosol particles and the inversion of multiwavelength data into microphysical particle properties.

Particles in the atmosphere have many different shapes. Therefore, Mie scattering theory is often a very rough approximation only. As long as the particles are small compared to the wavelength, the actual shape does not play a major role for the scattering properties as theories for non-spherical scatterers show. If the particles are large and non-spherical, like ice crystals, fluffy soot agglomerates, mineral dust, or sea-salt particles, we cannot use Mie scattering theory any more, but have to apply more elaborate non-spherical scattering theories. The presence of large non-spherical particles in the atmosphere can easily be detected with lidar. Spherical scatterers do not change the polarization state of linearly polarized laser light if scattered at  $180^\circ$ , whereas non-spherical scatterers lead to a depolarization of backscattered radiation. Polarization-sensitive light detection is particularly useful in the investigation of cirrus clouds and dust layers. The background and major findings of the polarization lidar technique are described in Chapter 2.

So far we assumed that each detected photon results from a single scattering process in the atmosphere. However, if the particle concentration is high and especially if the particles are large, as is the case in clouds, a photon can be scattered more than once before it reaches the lidar receiver. Again, the size of the particles plays an important role. Large particles show a strong forward-scattering peak due to light diffraction. Photons scattered at an angle close to  $0^\circ$  remain in the lidar's field of view, travel with the laser pulse, and can be backscattered (or the photon is backscattered and then undergoes one or several forward-scattering processes before reaching the detector). The effect of *multiple scattering* and how it can be corrected for or even exploited to provide information on cloud properties is described in Chapter 3.

Two special applications of elastic-backscatter lidar, the measurements of visibility and of cloud heights, are discussed in Chapter 6. These applications require comparably low instrumental effort and can routinely be used in traffic control, especially at airports.

A very specific form of an elastic-backscatter lidar is the high-spectral-resolution lidar described in Chapter 5. With an extremely narrow filter, realized by a Fabry–Perot etalon or an atomic-vapor or molecular-vapor absorption cell, the elastic backscatter signal from aerosol particles can be separated or removed from the molecular

backscatter component. The concept is based on the fact that, because of slow particle velocities, the spectral width of backscattering from particles is much narrower than the Cabannes line. Because the Rayleigh backscatter and extinction coefficients of the atmosphere can easily be calculated from pressure and temperature, the only unknown of a pure molecular backscatter signal is the particle extinction coefficient, which can thus be directly determined. The particle backscatter coefficient is independently inferred either from the signal transmitted by the Fabry–Perot etalon which contains virtually all of the aerosol, but only a small fraction of the molecular signal, or, in the case of the atomic or molecular cell filter, from the unfiltered signal. The same principle is used in the Raman lidar technique. Here, the molecular backscatter signal results from an inelastic Raman backscattering process from either nitrogen or oxygen as gases with known molecule number density in the atmosphere (see Chapter 4).

*Raman scattering*, as mentioned, is an inelastic scattering process which involves the change of the vibrational-rotational energy level of the molecule. The frequency shift of the scattered radiation corresponds to the energy difference between the initial and final molecular states and is thus specific for the interacting molecule. The change of rotational energy states leads to the rotational Raman side bands mentioned before. Because the population of energy levels follows Boltzmann's distribution law, the intensity distribution within the Raman bands contains information on the temperature in the scattering volume. One application of **Raman lidar** is therefore the measurement of atmospheric temperature profiles. This technique is described in Chapter 10. The change of the vibrational energy level results in frequency shifts of a few hundred to several thousand wavenumbers depending on the Raman-active molecule. Spectrally resolved analysis of backscattered radiation allows in principle the detection of a variety of atmospheric species. However, the comparably low Raman cross sections limit a meaningful use to gases present in relatively high concentrations. The Raman lidar technique is widely applied to the measurement of water vapor. The basic principles of Raman lidar, with special emphasis put on the observation of tropospheric water-vapor profiles, are explained in Chapter 9.

The detection of atmospheric gases with high sensitivity is possible with **differential-absorption lidar** or **DIAL**. The DIAL technique makes use of single absorption lines or broad absorption bands of gases. By emitting two wavelengths, one of which is absorbed more strongly than the other, the differential molecular absorption coefficient  $\Delta\alpha_{\text{mol,abs}}$

is determined. If the differential absorption cross section  $\Delta\sigma_{\text{mol,abs}}$  for the two wavelengths is known, the number concentration of the gas atoms or molecules can directly be deduced [see Eqs. (1.10)–(1.12)]. Chapter 7 introduces the application of DIAL for the measurement of gases such as  $\text{O}_3$ ,  $\text{NO}_2$ ,  $\text{NO}$ ,  $\text{N}_2\text{O}$ ,  $\text{SO}_2$ ,  $\text{CH}_4$ ,  $\text{HCl}$ ,  $\text{NH}_4$ , and others. DIAL is also of great interest for the observation of water vapor as the most important atmospheric greenhouse gas. Because of the narrow absorption lines of the  $\text{H}_2\text{O}$  molecule, water-vapor DIAL requires high stability and spectral purity of the emitted laser light and the consideration of Doppler broadening of the backscattered light. Thus, Chapter 8 deals especially with water-vapor DIAL. In addition, this chapter discusses the potential of DIAL for temperature profiling. The latter technique uses the temperature-dependent strength of absorption lines of oxygen; in this case the number concentration of the gas is known and the differential absorption cross section is measured which contains the temperature information sought.

If the two DIAL wavelengths are spectrally separated by more than just a few nanometers as in the case of ozone DIAL, differential backscattering due to the wavelength dependence of particle scattering becomes a major error source of this technique. The effect is hard to correct for if the particle scattering properties are not sufficiently well known. An alternative is the so-called **Raman DIAL**. Here, two nitrogen and/or oxygen Raman backscatter signals within the ozone absorption band are used to determine the ozone differential absorption coefficient. The differential backscatter coefficient is known in this case. The method is described as an application of Raman lidar in Chapter 9.

Two special forms of DIAL that may appear rather exotic even to long-term lidar practitioners are introduced in the final Chapter 14 of this book. BELINDA (for broadband-emission lidar with narrow-band determination of absorption), or “DIAL the other way around” as it is called by the authors, is based on the emission of laser radiation with a broader spectral width than the absorption line. The two DIAL signals are obtained by narrow-band filtering out portions of the backscattered light in the center and in the wings of the absorption line. With this approach the influence of line-broadening effects can be reduced. The price to pay is a large fraction of the backscatter signal lost in the filtering process. The second part of Chapter 14 describes white-light femtosecond lidar, a new and challenging approach of lidar. If the energy of a laser pulse is confined to a very short time interval of the order of femtoseconds, the pulse power can become as high as a few terawatts. At such high power,

qualitatively new interactions of the laser light with the surrounding medium occur. The non-linear Kerr effect leads to self-focusing of the laser beam in air, followed by plasma generation which in turn defocuses the beam and generates white light along the beam path. It was found that this white light predominantly travels in the backward direction, which makes it an excellent source for the use in lidar. On the way toward the receiver, the white light interacts with all atmospheric absorbers. Spectral analysis in the lidar receiver thus makes it possible to identify and quantify a multitude of atmospheric constituents. Even if a variety of technological and theoretical aspects have still to be solved and this technique cannot be applied to atmospheric research yet, it may give an idea of what kind the challenges of lidar research in the future may be.

Chapter 11 is the only chapter of this book that explicitly deals with the upper atmosphere. In the mesopause region, between about 80 and 110 km height, the presence of layers that contain metallic atoms and ions such as Na, K, Ca,  $\text{Ca}^+$ , Li, and Fe opened the field for **resonance fluorescence lidar**. Resonance fluorescence is obtained if the energy of the incoming photon coincides with the energy of a transition in an atom, ion, or molecule from one into another level. We speak of fluorescence because the reemission of light can occur at longer wavelengths. However, in this specific application the reemission is generally at the laser wavelength; therefore the technique is also called **resonance scattering lidar**. The extremely high cross sections for resonance scattering result in strong lidar signals and allow the determination of atom or ion number concentrations of less than  $10^8 \text{ m}^{-3}$  from distances of more than 100 km. In addition, the Doppler broadening and shift of the Na  $\text{D}_2$  line can be used to determine temperature and wind in this remote region of the atmosphere.

Turbulence and wind are the macroscopic manifestation of the collective motion of atmospheric molecules and particles. Its component along the line of sight of the laser beam Doppler-shifts the backscattered radiation to higher frequency if the scatterers move toward the lidar, and vice versa. By determining the frequency shift the wind speed along the lidar line of sight can be measured. The frequency shift is proportional to the ratio of wind speed and the speed of light and is thus extremely small. The detection of such small frequency shifts requires special instrumental efforts. **Coherent Doppler lidar** is based on the emission of single-mode single-frequency laser radiation and the coherent detection of the radiation backscattered from the moving particles. The return signal is mixed with the radiation from a local oscillator, and the frequency difference is

determined. In order to also determine the sign of the shift and not just its magnitude, heterodyne detection is applied, i.e., a frequency offset is introduced between the emitted laser pulse and the local oscillator. **Direct-detection Doppler lidar** uses the molecular backscatter component and measures the frequency shift by applying narrow-band spectral filters. Chapter 12 explains the Doppler lidar technique.

The application of lidar not just from ground, but from airborne and even space-borne platforms is of special interest for regional and global monitoring of atmospheric constituents. To reliably run active remote sensors on flying platforms is a great challenge. The effort, however, is rewarded by an incredible new insight into atmospheric processes, as only active remote sensing has the potential for high-resolution observations in space and time. The LITE (Lidar In-space Technology Experiment) mission on board the Space Shuttle in 1994 has proven the viability of lidar for this type of application and certainly was one of the milestones in lidar history. Chapter 13 reports on this interesting aspect of lidar research and discusses the prospects.

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## Polarization in Lidar

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### 2.1 Introduction

A fundamental principle of light is that the electric field  $\mathbf{E}$ -vector of the electromagnetic wave at any instant of time displays *some orientation* in space. This orientation can be fixed, yielding linearly polarized light, or rotating with time to yield circularly or elliptically polarized light. Random polarization is essentially a state in which a *beam* of light displays such a diversity of individual wave polarizations that no single state can be discerned with optical analyzers. Importantly, any state of polarization can be converted to any other state with the aid of a set of optical devices. Photons are pliable.

Historically, the discovery of the polarized nature of light evolved from experimentation with one type of optically active material, Iceland spar, which is a birefringent crystal of calcite that produces the phenomenon of double images. The dual images represent an image displacement during transmission through the crystal in two orthogonal polarization planes, and both Huygens and Newton demonstrated that this double refraction was intrinsic to a property of light and not the result of a modification induced by the crystal. Newton was unable to explain this phenomenon because of his adherence to corpuscular (light as a particle) theory, but, in his *Queries to the treatise Opticks*, hinted that double refraction represents an effect similar to the poles of a magnet. Thus, the term *polarization* was born. Further research led to the development of polarizing prisms by Rochon, Wollaston, Nicol, and my favorite for polarization lidar applications, by Glan. For a review of the development of the science of polarized light see [1].

Fortunately, as we will see, the pulsed lasers generally used in lidars naturally produce linearly polarized light because of the crystalline

nature of the lasing medium (e.g., a doped glass rod), and the method used in giant-pulsing, which typically relies on a polarization rotation device (e.g., a Pockels cell) to stop the cavity from lasing until the most propitious instant. Thus, the basic polarization lidar application involves the transmission of a linearly polarized laser pulse and the detection via a beam splitter of the orthogonal and parallel planes of polarization of the backscattered light. The ratio of these two signals, after adjustments are made to account for differences in the optical and electronic gains of the two channels, is referred to as the linear depolarization ratio, or  $\delta$  value. However, a variety of other measures of laser backscatter depolarization are possible, depending on modifications to the outgoing laser pulse and the number of polarization channels using various optical components.

Before going into greater detail, it should be mentioned that the polarization lidar technique was initially borrowed from analogous microwave radar methods developed mainly in the 1950s before the invention of the laser. Because of this, I will make references to the ground-breaking radar depolarization research. By the late 1960s, however, it became apparent that in comparison to microwave depolarization from nonspherical particles (typically smaller than the incident wavelength), laser depolarization (from particles larger than the wavelength) was considerably stronger, suggesting that polarization lidar had a promising future for the study of aerosols and the particles in clouds and precipitation (i.e., hydrometeors).

In the remainder of this chapter, I will discuss the types of depolarization measurements currently in use, explain the causes of laser depolarization in the atmosphere based on a combination of approximate theories and experiments, and provide examples of basic atmospheric research using this technique drawn mainly from our lidar research program. I will show that the lidar polarization technique greatly expands the capabilities of atmospheric probing with a variety of laser methods, and at a particularly economical cost in terms of extra components. In addition, as discussed in the final section, there remains a great potential for more advanced polarization lidar methods that no doubt will be fully explored in the not too distant future.

## 2.2 Measures of Depolarization and Their Uncertainties

As mentioned above, the workhorse of the polarization lidar field is the range( $R$ )-resolved linear depolarization ratio  $\delta$ , defined from [2] as

$$\delta(R) = [\beta_{\perp}(R)/\beta_{\parallel}(R)] \exp(\tau_{\parallel} - \tau_{\perp}), \quad (2.1)$$



where  $\beta$  and  $\tau$  are the backscattering cross sections and the atmospheric transmittances, respectively, in the planes of polarization orthogonal ( $\perp$ ) and parallel ( $\parallel$ ) to that of the laser. This definition comes from taking the ratio of the lidar equation in the two polarization planes, where most terms cancel out for each lidar shot. In practice, the exponential term is not used, but was originally included to account for the possibility that certain anisotropic targets like uniformly oriented ice crystals or raindrops could affect the transmission of light depending on the polarization state. Such effects are well known in microwave radar studies of precipitation, but have not been rigorously studied with lidars, which mainly have operated in the zenith direction at relatively short ranges. I will return to this topic later.

The general form of Eq. (2.1) represents the combined backscattering from (potentially) molecules  $m$ , aerosols  $a$ , and hydrometeors  $h$ , and so is sometimes referred to as the *total* linear depolarization ratio. This is because modern multichannel lidars based on advanced spectroscopic (Raman and high-spectral-resolution) techniques can intrinsically separate out the returns from molecules and aerosols, or aerosols plus hydrometeors, so it is possible to subscript  $\delta$  as  $\delta_m$ ,  $\delta_a$  in the absence of clouds, or  $\delta_{a+h}$ . Note, however, that the backscattering from hydrometeors typically dominates over that from molecules and aerosols, so that it is mainly in aerosol layers, where the backscattering contribution from air molecules can be similar, that the total linear depolarization ratio represents a mixture of atmospheric constituents [3].

Other measures of linear depolarization sometimes used are the range-integrated version from cloud (or layer) base to cloud top,

$$\Delta = \sum \beta_{\perp}(R) / \sum \beta_{\parallel}(R), \quad (2.2)$$

and the following form sometimes used in aerosol research

$$\delta'(R) = \beta_{\perp}(R) / [\beta_{\perp}(R) + \beta_{\parallel}(R)]. \quad (2.3)$$

Although rarely used in the lidar field, additional depolarization quantities exploited in radar research include the use of circular polarization (where in this case the parallel-channel backscatter is rotating in the opposite sense to that transmitted), combinations of linear and circular measurements, and differential reflectivity from lidars capable of transmitting and detecting both horizontally and vertically polarized light. Preliminary circular depolarization data have been reported from

cirrus clouds [4]. According to [5], the circular depolarization ratio  $\delta_c$  is related to the linear depolarization ratio by

$$\delta_c = 2\delta/(1 - \delta). \quad (2.4)$$

Backscattered laser light can also be evaluated with respect to the four Stokes parameters using a minimum of four receiver channels equipped with various linear and circular polarizing optics. Although some Stokes parameters have been tested in the field by lidar [6], to my knowledge a comprehensive analysis has only been attempted in the laboratory [7]. It was indicated in that study that the backscatter depolarization from ice crystal clouds consists of a combination of parallel-polarized and randomly polarized light. The parallel component represents fortuitous specular (mirrorlike) reflections off crystal faces, while the random part comes from the superimposition of numerous internal scattering events from a population of ice crystals displaying a variety of different shapes, sizes, and orientations. This has implications for understanding the possible errors in lidar  $\delta$  values.

Uncertainties in lidar depolarization measurements stem from various sources, but are basically related to errors in accounting for the differences in the optical and electronic gains of the channels, the polarization purity of the laser pulse, and the alignment between the polarization plane of the laser and that of the polarizer(s) in the detector(s). The simpler the design the better, and frequent calibration procedures should be performed. Figure 2.1 shows an early but still common type of receiver design that incorporates a collecting lens (instead of the telescope mirror assembly used in lidar), a laser line interference filter, pinhole aperture, Glan-air polarizing prism, and dual photomultiplier tubes placed at the  $108^\circ$  polarization separation angle [8]. We have suggested in [9] applying two corrections to the lidar signal strengths (or power)  $P$  according to

$$\delta(R) = [P_\perp(R)/P_\parallel(R)]K - \chi, \quad (2.5)$$

where the calibration constant  $K$  accounts for the differences in the entire detector channels obtained by viewing an unpolarized light source, and  $\chi$  is a correction term to account for any slight mismatch in the transmitter and detector polarization planes plus any impurity in the laser polarization state. The correction factor can be estimated by monitoring the  $\delta$  values in the middle and upper troposphere, where the effects on depolarization of aerosols are normally small. Note that in some lidar systems a rotating quarter-wave plate is used to bring the receiver into

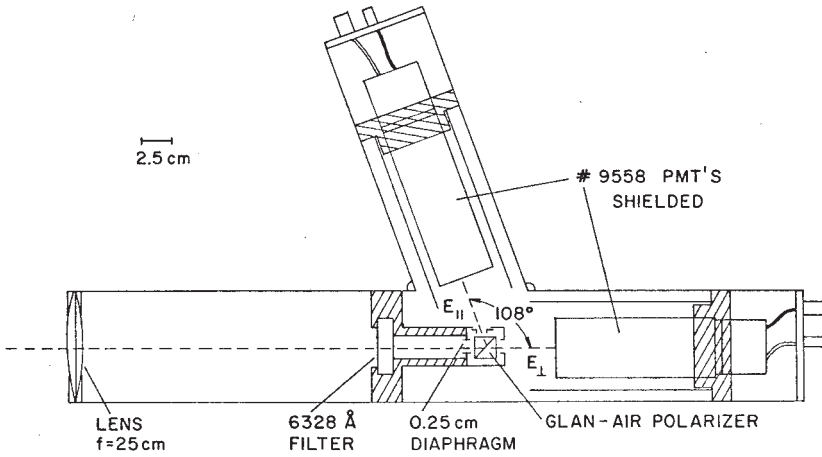


**Fig. 2.1.** Design of a dual-channel polarization receiver used in early laboratory studies of the backscatter laser depolarization technique from [8], but analogous to modern lidar designs. For the detection of the  $0.6328\ \mu\text{m}$  laser light, two photomultiplier tubes (PMTs) were used.

proper alignment, but a simple, well-machined design is preferable for most applications. The basic considerations in polarization lidar design are discussed in [10].

Finally, careful attention should be given to appropriate signal-processing and -averaging approaches to minimize the effects of signal noise, without overaveraging to lose the often detailed structures of atmospheric targets.

### 2.3 Causes of Lidar Depolarization: Approximate Theories

In the pioneering polarization lidar research reported over 30 years ago in [2], it was clear that a new horizon was opening into the characterization of atmospheric particles. Its basic utility is rooted in various scattering theories. According to the exact Lorenz–Mie theory, spherical particles that are homogeneous in content (with respect to the refractive index) always backscatter linearly polarized electromagnetic radiation in the same (incident) plane of polarization. A variety of approximate scattering theories predict that nonspherical or inhomogeneous particles will introduce a depolarized component into the backscattering. Thus, polarization lidar is unique among remote sensors in that it has the potential to unambiguously identify the thermodynamic phase of clouds. The

strength of the depolarization process in nonspherical particles depends on the amount and complexity of the particles' deviation from spherically symmetrical shape, but also on the particle size relative to the wavelength (as expressed in the size parameter  $x = 2\pi r/\lambda$ , where  $r$  is the particle radius and  $\lambda$  the incident wavelength) and the particle refractive index at  $\lambda$ .

In considering the differences between radar and lidar backscattering from nonspherical hydrometeors, the usual radar case is described adequately by the Rayleigh–Gans theory using spheroidal particle models for  $x \lesssim 0.1$ – $0.4$  [11], whereas at greater  $x$  a set of theories is necessary to describe the general situation for lidar. For infinitely large nonspherical particles in the geometric optics domain (in practice for  $x \gtrsim 50$ – $100$  according to [12]), scattering is described by ray-tracing theory that implicitly treats depolarization by resolving the rotation of the incident **E**-vector according to the laws of optics through those series of internal refractions and reflections that result in backscattering. Figure 2.2 schematically shows the differences in how the backscattering from spheres and ice crystals can be treated [13]. This general ray-tracing approach has long been used to explain the presence of halos and arcs from hexagonal ice crystals suspended in the atmosphere. However, the exact particle shape is of great importance in these intensive computer computations, so the realism of the model shape has a significant influence on the applicability of the  $\delta$  value predictions. The calculations are normally based on pristine hexagonal ice crystal shapes, but such models fail to treat the diversity of ice crystal shapes found in nature. Suggested solutions to this problem involve the use of hybrid particle shapes such as fractal or Chebyshev particles, which, although clearly unrealistic, may on average mimic the scattering properties of an ensemble of particles that display a wide variety of hexagonal shapes and orientations [14].

For nonspherical or inhomogeneous particles of a size comparable to the incident laser wavelength, such as newly formed ice crystals or aerosols with inclusions, other scattering theories continue to be developed. These theories must essentially cover the Rayleigh–Gans to geometrical-optics transition zone, and include the discrete dipole approximation [15], the T-matrix approach [16], and the finite difference time domain method [17]. These approximate theories are believed to yield reliable results for  $x \lesssim 15$ ,  $x \lesssim 100$ , and  $x < 15$ – $20$ , respectively. In terms of Lorenz–Mie theory, the  $x$  domain between  $\sim 5$  and  $40$  is referred to as the resonance region because of the large variations in

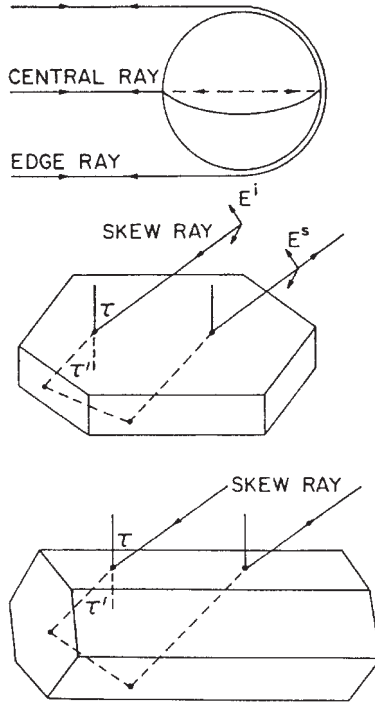


**Fig. 2.2.** Rendering of the incident ( $E^i$ ) and backscattered ( $E^s$ ) waves from a spherical drop and two (plate and column) hexagonal ice crystal models using the ray-tracing approach, where  $\tau$  and  $\tau'$  are the incident and refracted skew ray angles [13].

scattering parameters found with changing sphere size. For an evaluation of the dependence of the generation of depolarization on ice particle size using the T-matrix approach, see [18]. It was shown in that study using various nonspherical particle models that  $x > 5$ –10 are needed in order to generate the  $\delta$  values typical of those in the large-particle limit.

Finally, the refractive index also influences the amount of depolarization generated by nonspherical particles. Laser backscatter depolarization is essentially confined to those nonspherical particles that do not have overwhelming absorption at the laser wavelength, because depolarization results predominantly from internal reflections. For water and ice particles, and most aerosols, only visible and near-infrared Nd:YAG ( $1.06\text{ }\mu\text{m}$ ) laser wavelength lidars will easily detect depolarization. Midinfrared  $\text{CO}_2$  ( $\sim 10.6\text{ }\mu\text{m}$ ) lidars, on the other hand, will not measure significant  $\delta$  values in ice clouds because the

strength of the ice absorption process is so dominant: slight  $\delta$  values apparently occur due to a form of multiple scattering from surface reflections between the facets of complex ice crystals [19]. Below, when I refer to polarization lidar, it is implicit that I am referring to those with laser wavelengths for which particle absorption is not overwhelming.

In practice, it should be stressed that assemblies of spherical particles in water droplet clouds can produce non-negligible depolarization because of multiple scattering activity in the finite lidar field-of-view, or FOV [20]. Typical lidar FOVs of a few milliradians promote this effect, although in lidars using FOVs on the order of 0.1 mrad this influence can be reduced to the point of being negligible. Moreover, the tendency for certain ice crystal shapes to orient uniformly in space with their maximum dimensions parallel to the ground can result in ambiguous  $\delta$  values: most commonly horizontally oriented plate ice crystals are observed to produce non-depolarizing specular reflections in the zenith-pointing direction [21]. This anisotropy, however, is easily recognized by pointing the lidar a few degrees off the zenith direction [22]. Thus, to ensure unambiguous cloud phase discrimination, it is important to have at least limited (near-zenith) scanning capabilities of the lidar table [23].

## **2.4 Lidar Depolarization in the Atmosphere**

In this section I discuss the generation of lidar depolarization according to the nature of the atmospheric target. This assessment is based on over 30 years of lidar field measurements and lidar scattering simulations.

### **2.4.1 Pure Molecular Scattering**

Because the sizes of typical molecular species are very much smaller than lidar wavelengths, to lidar the molecular atmosphere is a Rayleigh scattering environment, and considerable backscattered signal is measured with near-ultraviolet and visible lidars. Hence, lidar depolarization from the molecular atmosphere may be calculated [24]. As also shown by experiments, molecular  $\delta$  values are typically on the order of a few percent, and can be neglected in most clouds. Precise knowledge of molecular depolarization at the lidar wavelength can assist in calibration and the identification of multiple scattering effects in the case of spectroscopic lidars.

### 2.4.2 Aerosol Scattering

A variety of particles, both dry and wet (i.e., deliquesced), can be found suspended in the atmosphere. Aerosol types include haze, wind-risen dust, smoke, volcanic emissions, particles released through pollution (e.g., carbon-based) or by the surface of the ocean, and those created by gas-to-particle conversions. Their sizes vary a great deal, from molecular cluster-sized when newly formed to particles of several microns dimension, which have limited lifetimes due to an appreciable sedimentation rate. This rich tapestry of atmospheric aerosol conditions presents both challenges and opportunities for polarization lidar research. In an earlier review of polarization lidar in atmospheric research [10], it was pointed out that applications to aerosol research were previously underappreciated: recent lidar research directions are correcting this situation.

Because of the great range of aerosol sizes, they span the region between the Rayleigh- and geometric-optics-scattering domains. Many aerosols consist of spherical particles, such as deliquesced aerosols, volcanic sulfuric acid droplets, and spume drops released by the action of wind on water waves. Little or no lidar depolarization can be expected from these targets as long as they are reasonably homogeneous. For irregularly shaped aerosols, particularly volcanic and desert dusts, and markedly inhomogeneous particles like partially crystallized acid droplets, the amount of depolarization measured will depend strongly on the size parameter  $x$  [18], and also to some degree on the refractive index at the laser wavelength. Although little depolarization is expected from minute or strongly absorbing (e.g., carbon-black) aerosols, unfortunately even the largest particles are too small to be accurately treated by ray-tracing theory to predict lidar  $\delta$  values. Lidar data indicate, however, that supermicron-sized desert dust clouds generate  $\delta$  up to  $\sim 0.25$  [25, 26], in contrast to the near-zero values measured in haze [27].

### 2.4.3 Clouds of the Middle and Upper Atmosphere

An area of active polarization lidar research involves the ground-based and airborne study of various types of polar stratospheric clouds (PSCs), which often more closely resemble aerosol layers than proper clouds because of the minute particle sizes and exotic chemistry involved at these frigid temperatures. As a matter of fact, perhaps more than any other application, our fundamental understanding of these targets has largely been shaped by airborne polarization lidar observations in polar

regions. The study of these rare clouds has gained importance because of their connection to stratospheric ozone depletion during the polar winter.

As reviewed recently in [28], PSCs can be composed of mixtures of water, sulfate, and nitric acid solutions, and occur in both the solid and liquid phases. At least two basic types appear to exist. Type I PSCs are found somewhat above the frost point of ice (typically at  $\sim -85^\circ\text{C}$ ) and have been broken into two subtypes. Type Ia display higher depolarizations than type Ib, which are assumed to be small liquid particles composed of supercooled sulfuric-acid ternary solution (STS), an aqueous solution of sulfuric and nitric acids. The higher  $\delta$  in the former case indicate small solid particles perhaps composed of nitric acid trihydrate (NAT). Type II PSCs, on the other hand, occur at colder temperatures and generate strong depolarization consistent with ice crystals. Although in some cases mixed particle conditions may occur to complicate the analysis, it is clear that polarization lidar offers considerable promise in characterizing these exotic clouds [3]. Because PSC particle size is often close to the common lidar wavelengths, multiwavelength lidar depolarization techniques are especially well suited [29, 30].

Finally, even higher in the atmosphere, at  $\sim 80$  km, reside the rare noctilucent clouds (NLC) that are seen mainly in polar regions. It has been assumed that they consist of minute ice particles, and recent polarization lidar data confirm this suspicion. As reported in [31], although the cloud depolarization is low (i.e.,  $\delta = 0.017 \pm 0.01$ ), it is sufficiently nonzero to indicate nonspherical particle shapes. There is evidence that NLC are increasing, perhaps an indication of the effects of a change of global climate.

#### 2.4.4 Water Cloud Scattering

Analysis of Lorenz–Mie theory demonstrates that the mechanisms responsible for laser backscattering under the spherical symmetry assumption involve only front- and rear-surface axial reflections, and surface waves from light that gets trapped at the dielectric interface. These mechanisms fail to produce laser depolarization in the backscatter from a single particle, but Lorenz–Mie theory can be used to explain why lidars can measure significant amounts of linear depolarization in water clouds containing populations of spherical cloud droplets [32].

The signature of this process is the steady increase in depolarization as the laser pulse penetrates into the cloud. This has long been known to



be a result of photon multiple scattering activity in the dense assemblage of cloud droplets, with concentrations typically measuring in the hundreds per cubic centimeter. Theoretical simulations have shown that the strength of this process depends on the droplet concentration, the distance to the cloud (i.e., the lidar footprint), and, operationally, on the size of the detector FOV. Depolarization is a byproduct, as variable FOV lidar research in water clouds has clearly shown. The source of the depolarization is laser light mainly scattered into the near-backward direction in certain azimuthal planes, as revealed by Mie theory [32]. This light becomes depolarized with respect to the incident plane, and if redirected into the lidar receiver by second- or higher-order multiple scatterings, a mixture of non-depolarized primary and multiply scattered light is detected. Depolarization increases with cloud depth because the effect of the primary scattering decreases as the laser pulse attenuates, while multiple scattering accumulates. The change in the size of the growing cloud drops above cloud base also has a strong effect [33].

#### 2.4.5 Ice Cloud Scattering

Ice phase clouds, principally the varieties of cirrus clouds that inhabit the upper troposphere, contain, in comparison to water droplet clouds, large and decidedly nonspherical particles that can be modeled via ray-tracing theory. The exceptions involve particularly frigid cirrus clouds, which contain crystals small enough ( $\lesssim 30 \mu\text{m}$ ) to generate solar corona [34], and even smaller crystals in aircraft condensation trails, or contrails. Lidar  $\delta$  in young contrails appear to vary widely, but no evidence for spherical or near-spherical particles has been found [35, 36]. Other lidar data indicate that even aged ( $\sim 1$ -h old) contrails contain particles so small that pronounced differences in  $\delta$  values at the 1.06 and  $0.532 \mu\text{m}$  Nd:YAG wavelengths have been measured [37]. Such targets are an exception to the rule for ice clouds, however. The influence of contrail particle size on depolarization is discussed in [18].

Ray-tracing calculations based on pristine hexagonal ice crystal shapes indicate that the  $\delta$  values for randomly oriented ice crystals tend to increase as the particle axis ratio increases (i.e., from plates to columns), as shown in Table 2.1. The  $\delta$  values even for thin plate ice crystals, however, are nonetheless large compared to other atmospheric targets. Although this may seem to be a useful finding for inferring the composition of ice clouds, cirrus ice crystals sampled *in situ* often show hollow, complex spatial, and irregular or rounded shapes. Moreover, the normal

**Table 2.1.** Backscatter linear depolarization ratios<sup>a</sup>

| $L/2a$               | $\delta$ | $\delta_b$ |
|----------------------|----------|------------|
| 8/80 (thin plate)    | 0.339    | 0.399      |
| 16/80 (plate)        | 0.355    | 0.396      |
| 32/80 (thick plate)  | 0.394    | 0.508      |
| 64/80 (short column) | 0.382    | 0.500      |
| 200/80 (column)      | 0.550    | 0.616      |
| 400/80 (long column) | 0.563    | 0.611      |

<sup>a</sup>Predicted by ray tracing for randomly oriented solid ice crystals with the indicated length  $L$  to radius  $a$  axis ratios (in  $\mu\text{m}$ ). Results computed ignoring ( $\delta$ ) and including ( $\delta_b$ ) ice birefringence effects.

situation seems to involve a diverse mixture of ice crystal types caused by a combination of physical cloud processes including new ice crystal nucleation, vertical transport, and ambient growth/evaporation conditions. An exception seems to involve cirrus clouds that produce brilliant halos, which characteristically generate relatively low  $\delta$  indicative of thin plate crystals [38].

Climatological findings using polarization lidar reveal that  $\delta$  steadily decreases with increasing height or decreasing temperature in cirrus clouds [9, 39]. This is not a consequence of photon multiple scattering, however, for even optically and physically thin ice clouds at cold temperatures generate relatively large  $\delta$ . Rather, this finding is believed to reflect the gradual change in basic ice crystal shape, from plates to columns, with decreasing temperature, along with such other factors as solid versus hollow particle effects and changes in their ability to orient in the horizontal plane. The depolarization data also show that the presence of supercooled water droplets in cirrus is uncommon, mainly restricted to transient patches near relatively warm cirrus cloud bottoms [40].

Moreover, the sensitivity of polarization lidar to particle phase and shape is so great that in recent years it has provided rare evidence for indirect climate forcing from the effects of aerosols on clouds. Unusually high  $\delta$  were found in a cirrus cloud along a tropopause fold following the 1991 Pinatubo volcanic eruption: the high  $\delta$  were attributed to changes in ice crystal shape following nucleation from sulfuric acid droplets [41]. Similarly, unique lidar depolarization scan data and cirrus optical displays were noted in midlatitude cirrus derived from tropical thunderstorm outflow, suggesting the effects of sea salt or some other form of marine nuclei on ice crystal structure [42]. Finally, cirrus clouds studied far downwind of Asian dust storms were found to have unusually

warm temperatures, which was suggested to be a result of the strong ice nucleating capabilities of desert clay particles in turning supercooled water clouds to ice [26].

#### 2.4.6 Mixed-Phase Clouds

It is in the study of mixed-phase clouds that the unique capability of polarization lidar to identify cloud phase is of crucial importance. In the presence of the ice particles that typically trail below supercooled mixed-phase clouds, a phenomenon called *virga*, even the best (shortest-wavelength) microwave radar measurements would detect mainly the larger, precipitating particles within and below the cloud. This is also true for the case of rain or snow reaching the ground beneath the base of the source cloud (see below). Although the depth of lidar probing is restricted in optically dense targets, and may only be a few hundred meters into the source cloud, lidar has the ability to locate accurately the liquid cloud base position using depolarization data. As a matter of fact, the use of polarization lidar in field experiments first became prominent during weather modification research in winter mountain storm clouds, which also saw the early use of microwave radiometers and millimeter-wave cloud radars [43]. Because each type of remote sensor has its advantages and disadvantages in cloud remote sensing, their coordinated use, termed the multiple remote sensor approach, is still at the foundation of modern field experiments and attempts to identify cloud type [44].

Interestingly, ice crystal growth in supercooled liquid clouds often seems to favor large plate crystals, which produce near-zero  $\delta$  values when horizontally oriented in and below the cloud [40]. Not to be confused by this, it is again useful to have the ability to scan the lidar off the zenith direction. More research is needed to determine if the  $\delta$  values measured in the multiple-scattering-dominated medium of the mixed-phase cloud itself can be used to quantify, or even reveal, the presence of the ice phase constituents.

#### 2.4.7 Precipitation Scattering

Precipitation reaching the ground can be grouped into three main categories: snow, rain from melting snow, and rain or drizzle that has formed through the coalescence process (without the intervention of the ice phase). Polarization lidar, under the proper laser-transmissive conditions, provides useful data to identify these precipitation generating mechanisms, and much more.

Snowfall can be comprised of ice crystals of various shapes, their aggregates, or rimed particles like graupel. Lidar  $\delta$  values can be used to discriminate between rimed and unrimed particles, according to some field studies [23], because the frozen cloud droplets can increase the complexity of the particle shape. When ice crystals of various shapes aggregate into snowflakes, relatively strong depolarization generally appears to result, and complex-shaped radial ice crystals may also generate the same results.

That rain at the surface often began as falling snow has long been known, but the changes that occur in the backscattering of microwaves during the phase transition later became a curiosity to radar meteorologists, and still later, to polarization lidar researchers. Indeed, understanding the effects on scattered electromagnetic energy of the complex changes in hydrometeor shape, phase, refractive index, and how these depend on cloud microphysical conditions, is a harsh test for any scattering theorist or cloud modeler. The chief consequence of this combination of factors at microwaves is the radar bright band, a fairly narrow feature of peak radar reflectivity factors that occurs, approximately, where wet snow changes to rain. A peak in radar linear depolarization also occurs at about this position in the melting layer [45]. These features appear to result from refractive index and particle shape, size, and fallspeed effects. The case at visible wavelengths, on the other hand, is quite different when the laser pulse can penetrate into the melting region without completely attenuating. An analog to the radar bright band occurs due to the strong attenuation often noted in the snow above the melting region, a quite different cause from the refractive index change using radar. A lidar depolarization bright band occurs under some conditions, which appears to result from the complex shape of partially melted snowflakes [46]. The most interesting phenomenon is called the lidar dark band [45], which appears to occur where severely melted snowflakes collapse into mixed-phase raindrops, and so involves depolarization to some extent because of the optical inhomogeneity within the drop.

Rain itself, regardless of the generating mechanism, is composed of distributions of drops whose departures from sphericity depend on fall velocity, or raindrop diameter. The balance between the force of the drops' surface tension and aerodynamic drag forces determines the exact form of the shape distortion. Since these distortions begin to become non-negligible for drop diameters  $\gtrsim 100 \mu\text{m}$  in the lower troposphere, it is only in drizzle that lidar  $\delta = 0$  are possible in dilute assemblies of such drops. Typical millimeter-size raindrops can be expected to induce some

backscatter depolarization, although laboratory studies with artificial raindrops show only slight depolarizations [23]. However, it is also indicated that significant  $\delta$  occurred in single drops that grazed obstructions during fall and thus underwent strong shape oscillations or breakup, such as might occur from drop collisions or the effects of turbulence in the atmosphere. Because these findings come from laboratory studies, which probed the drops at horizontal incidence, it can be questioned how applicable the results are to raindrops sampled by zenith or scanning lidars. Certainly, scanning lidar measurements have indicated sometimes dramatic backscattering anisotropy in rainfall [47, 48].

## 2.5 Notable Applications in the Field

In this section I present some examples of polarization lidar data from clouds and aerosols to illustrate the technique's basic utility in distinguishing spherical from nonspherical scatterers. The lidar used to provide most of these examples is the Polarization Diversity Lidar (PDL), which was designed to be a testbed of lidar polarization techniques [37]. This mobile system is based on a 10-Hz Nd:YAG laser transmitter with dual outputs of  $\sim 0.35$  J at the  $1.06\text{ }\mu\text{m}$  and frequency-doubled  $0.532\text{ }\mu\text{m}$  wavelengths. The laser is mounted on a fully scannable lidar table that also supports two telescope receiver units, a video camera, and a safety radar laser-shutdown device to avoid accidental irradiation of aircraft. Data digitization at 10 Hz can be accomplished down to a range resolution of 1.5 m. Dual-wavelength linear depolarization data are normally collected, one telescope for each color. However, the receiver units are interchangeable and additional measurement techniques can be quickly applied: a nitrogen Raman channel is currently being added, and plans include additional Stokes parameter measurements.

The first two cases come from the July 2002 Cirrus Regional Study of Tropical Anvils and Cirrus Layers–Florida Area Cirrus Experiment (CRYSTAL–FACE) field campaign that was conducted in southern Florida to research subtropical thunderstorms and the cirrus that evolve out of thunderstorm anvils [49]. The PDL was located in the western Everglades at Ochopee. In these examples are shown height-versus-time zenith-lidar displays of the linear depolarization ratio (see color  $\delta$ -value key at top) above a grayscale of lidar attenuated backscattering (based on the logarithm of the signal strength) from the  $0.532\text{ }\mu\text{m}$  channels. The PDL data in Fig. 2.3 show an unusual event that is uniquely captured

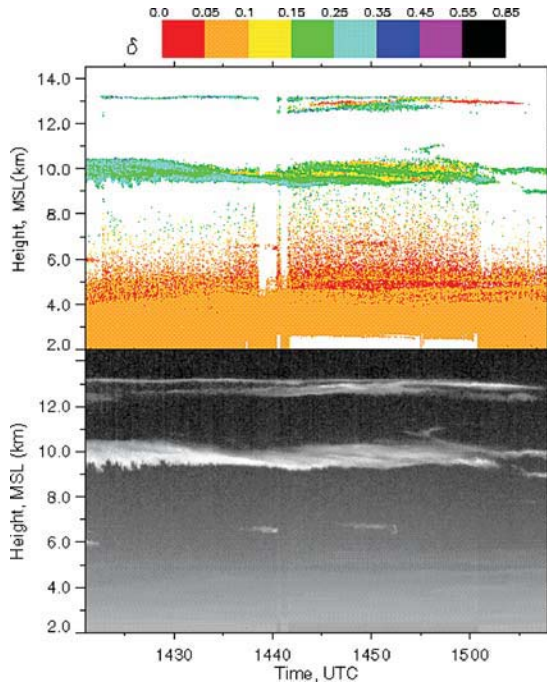


**Fig. 2.3.** Polarization lidar height-versus-time displays of range-normalized, attenuated, parallel-polarized backscattered signals (bottom, using a logarithmic gray scale) and linear depolarization ratios (see color  $\delta$ -value scale at top) from the  $0.532\ \mu\text{m}$  channel of the Polarization Diversity Lidar (PDL), collected on 29 July 2002 from Florida's western Everglades during the CRYSTAL-FACE campaign [49]. Depicted are two upper-tropospheric ice clouds, and a more weakly depolarizing layer of nonspherical dust particles below  $\sim 5.5$  km that was transported across the mid-Atlantic Ocean following a Saharan dust storm.

by polarization lidar. Present in the upper troposphere are two cirrus cloud layers generating, mostly, strong depolarization. The upper layer is located near the tropopause and probably represents a variety of optically subvisual cirrus believed to be widespread in the tropical and subtropical regions. In this cold cloud is a relatively strongly backscattering layer of low ( $<0.05$ )  $\delta$  values, indicating the presence of horizontally oriented plate ice crystals. The lower cirrus layer centered at 10 km height appears to be a remnant of an anvil advected in from thunderstorms developing to the north of the site. Depolarization ranges from 0.1 to 0.4, and at cloud base near the beginning of the display are structures that are the remnants of anvil mammata. Of greater interest, however,

is the evidence below  $\sim 5.5$  km for desert dust transported across the mid-Atlantic Ocean following a Saharan dust storm. (Note that  $\delta$  values cannot be calculated generally below 2.0 km in this case because of strong off-scale signals.) The maximum 0.10–0.15  $\delta$ -values in the aerosol are somewhat lower than the  $\sim 0.25$  value typical of many Asian dust storm particles [26], but they are still much higher than the usual (marine haze) boundary-layer aerosol with near-zero  $\delta$  [27]. In this case, the lidar returned power display reveals that the aerosol scattering is barely discernable, so the  $\delta = 0.10$ –0.15 represent a dilute mixture of the aerosols with air. As described elsewhere [49], backtrack trajectory and chemical aerosol analyses confirm that the origin of the aerosol was from the Saharan region.

An expanded display of a supercooled liquid altocumulus cloud is shown in Fig. 2.4. These  $0.532\text{ }\mu\text{m}$  data were collected immediately after

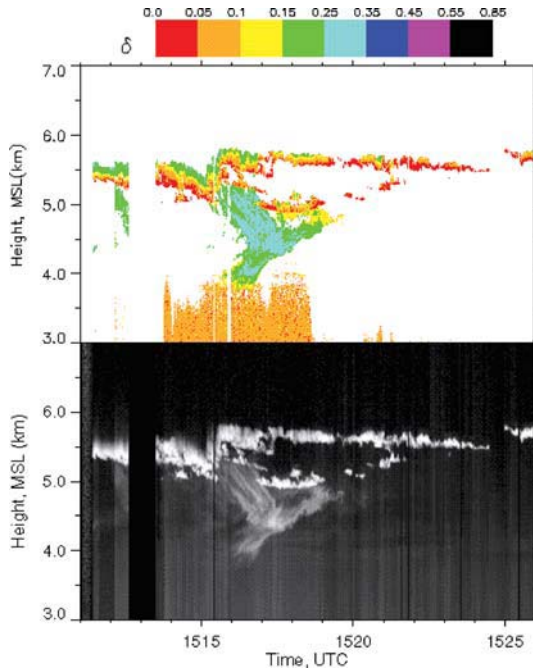


**Fig. 2.4.** PDL displays of a mildly supercooled liquid altocumulus cloud layer sampled immediately after the period shown in Fig. 2.3. The cloud at times shows strong multiple-scattering-induced depolarization, and also a brief glaciation that produced an  $\sim 1$ -km deep fallstreak of strongly depolarizing ice crystals. Contact with the top of the dust layer likely has produced this relatively warm cloud phase change [49].

those in Fig. 2.3, but because the detector sensitivity was lowered to best observe the relatively strongly backscattering liquid-dominated layer, much of the aerosol information was lost (but can be assumed to be the same as in Fig. 2.3). (Note that the gaps in the data record correspond to the passage of cumulus and fractus clouds, which blocked lidar probing at the boundary layer top.) The liquid cloud base position can be identified by the start of strong backscattering and the corresponding near-zero  $\delta$  between about 5.0 and 6.0 km. Initially the layer was dense enough to completely attenuate the lidar pulses and generate significant multiple-scattering-induced depolarization. But as the layer descends, it bifurcates and briefly glaciates at  $\sim 1516$  UTC: note the 0.3–0.4  $\delta$  values from ice crystals descending well below the layer in the virga. Subsequently the layers gradually thinned while the altocumulus  $\delta$  values decreased and the upper cirrus layers were again detected. What is of particular interest is that the liquid cloud phase transition, at a relatively warm temperature of  $\sim -5$  to  $-9.0^\circ\text{C}$ , corresponds to the height shown earlier to have been the top of the depolarizing aerosol. Thus, it appears that the ice nucleating abilities of Saharan mineral dust are similar to those of its Asian counterpart [26].

Scanning PDL data can reveal more intricate details concerning the shapes and orientations of ice crystals, as is shown in Fig. 2.5. The data were obtained at the Facility for Atmospheric Remote Sensing (FARS) in Utah, US, from an unusual cirrostratus cloud of tropical origin, which produced vivid optical displays including a rare Parry arc [42]. The two  $\pm 10^\circ$  range-height scans from the zenith direction were obtained during periods when the Parry arc was absent (left) and especially pronounced (right). The lidar scan on the left corresponds to a cirrus cloud containing a mixture of horizontally oriented plate crystals with those of different shapes and/or orientations, which produced relatively high depolarization (compared to the near-zero zenith  $\delta$  in Fig. 2.3) and a weak and irregular backscattering enhancement in the zenith. The dissimilar scan on the right, on the other hand, is unique in my experience. Although lower  $\delta$  again occur within about  $\pm 1.0^\circ$  of the zenith, between  $\sim 1.0$  and  $2.0^\circ$  from the zenith, the depolarization is extremely high and approaches unity. It was speculated that this highly anisotropic depolarization pattern was caused by the dominant ice crystal shape and orientation responsible for the Parry arc, as was supported by ray-tracing simulations using the so-called Parry arc column crystal orientation. Such crystals fall with a pair of prism faces closely parallel to the ground in a rare form of three-dimensional aerodynamical alignment.



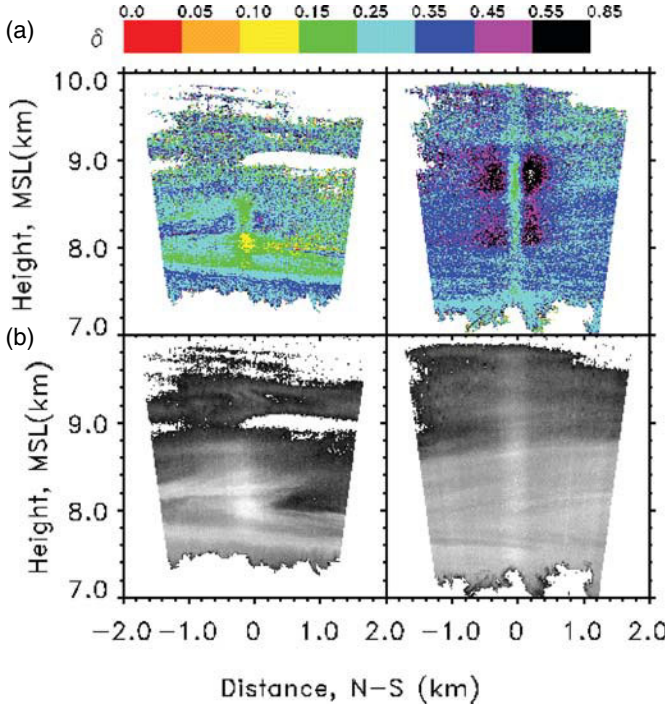


**Fig. 2.5.** PDL backscattering (b) and depolarization scans (a) collected  $\pm 10^\circ$  from the zenith direction to study the ability of ice crystals to orient horizontally in space. Data collected in the green ( $0.532 \mu\text{m}$ ) channel on 16 November 1998 at the Facility for Atmospheric Remote Sensing (FARS) in Salt Lake City, UT, from a cirrus cloud layer that produced exceptional halos and arcs including the rare Parry arc [42].

Figure 2.6 depicts peculiar lidar anisotropic backscatter and depolarization effects in rain over almost full  $180^\circ$  range-height scans from a field site north of Montreal. The polarization data were obtained with a  $0.532 \mu\text{m}$  laser transmitter and a rotating mirror scanner [47]. In each case are shown in (a) the relative returned (attenuated) laser power and linear depolarization ratios (note color scales at right of the elevation angle scans), and in (b) the angular dependence of the total orthogonal ( $s$ ) plus parallel ( $p$ ) returned powers (blue) and  $\delta$  (i.e.,  $s/p$ ) values (red) averaged over 500-m height intervals in the rain. In the left column, the returned power scan data reveal a strongly scattering and attenuation-limited snowfall layer overlying the lidar dark band [45], with rain below. The corresponding depolarization pattern suggests more intimate details. The relatively low zenith  $\delta$  in the ice/snowfall layer indicate the presence

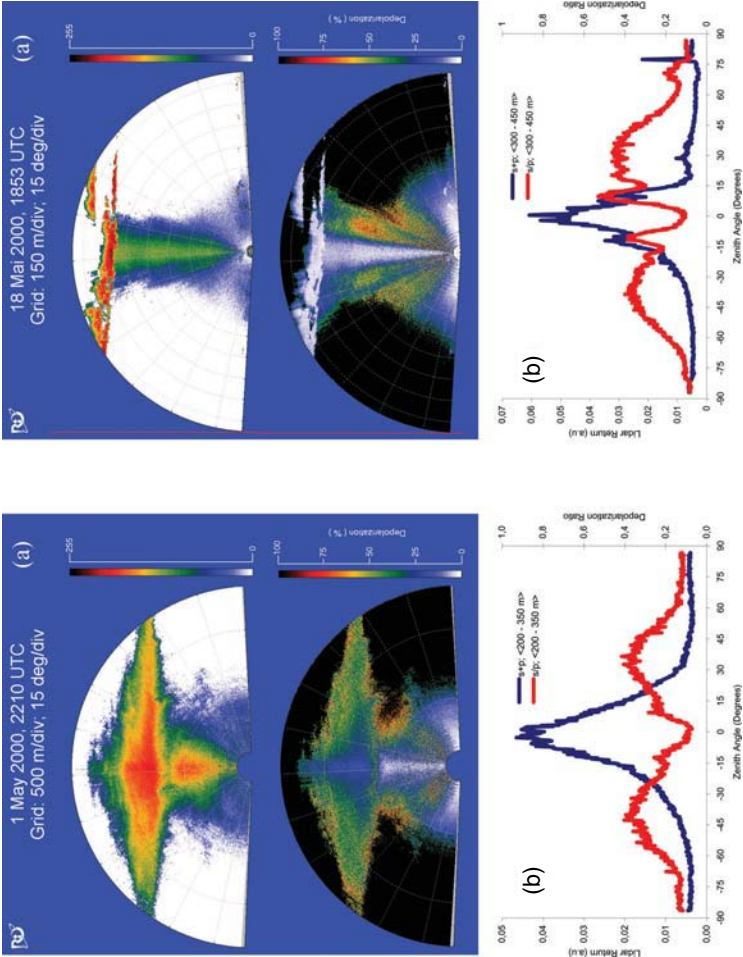


**Fig. 2.6.** Panels (a) at the top show almost full ( $180^\circ$ ) range-height attenuated backscattering (in arbitrary units) and depolarization scans obtained in rainfall on 1 and 18 May 2000 with a  $0.532 \mu\text{m}$  lidar from a site north of Montreal, Canada, just south of the Laurentian Hills [47]. In panels (b) at the bottom are shown (in blue) the total backscattering and (in red) the linear depolarization ratios averaged in the rain. The odd patterns in the backscattering and  $\delta$  value displays appear to be a result of sampling aerodynamically distorted oblate raindrops.

of some horizontally oriented planar ice crystals, which, interestingly, seem to wobble from the horizontal plane more widely as particle melting begins just above the dark band. The strongest zenith  $\delta$  occur in the dark band zone, where severely melted particles no longer have the ability to orient uniformly. Below, the near-spherical raindrops generate quite low  $\delta$  in the zenith, as one would expect, as well as strong backscattering. However, in (b) the  $\delta$  in the rain display peaks at about  $\pm 40^\circ$ , which must reflect the effects of aerodynamically distorted raindrop shapes. This, however, remains to be rigorously investigated.

In the right-hand column of Fig. 2.6 is shown a pure-rain situation below a liquid cloud layer that displays intense (attenuation-limited) backscattering and near-zero  $\delta$  at and near the cloud base, with multiply scattered depolarization increases aloft. In this case the light rainfall generates strong backscattering only surrounding the zenith direction. The  $\delta$ -value scan, on the other hand, shows a complex pattern with a minimum near the zenith surrounded by two peaks that seem to defy explanation. I conclude that such a strange pattern in  $\delta$  reflects the peculiarities of the size-dependent raindrop shapes sampled at this time, although it is also possible that the corrections needed to adjust the  $\delta$  using a mirror have not fully accounted for the backscattering properties of oddly shaped raindrops capable of causing strange elliptically polarized patterns.

## 2.6 Outlook and Conclusions

There is little doubt that the laser backscatter depolarization technique used by polarization diversity lidar has unique cloud and aerosol research capabilities that have already contributed significantly to our knowledge of the state of the atmosphere. Major applications have involved identifying supercooled liquid cloud layers in storms, the study of ice crystal shapes and orientations in cirrus clouds, and characterizing the nature of polar stratospheric clouds. In addition, this technique also shows promise for discriminating between basic aerosol types and identifying the indirect effects of aerosols on clouds because of its unique sensitivity to both of these species [26]. Polarization lidar applications were among the first for lidar both from the ground [2] and from aircraft [50]. Polarization lidar today is undergoing a renaissance in supplying useful and synergistic data with routine applications to aerosol lidars, differential absorption, high spectral resolution, and Raman lidars. As aforementioned, I stress that the additional findings are obtained economically. Moreover, as

a component of multiple remote sensor studies, polarization lidar is a vital complement for a variety of applications.

New laboratory and theoretical studies should also benefit the field. However, due to the propensity for large nonspherical hydrometeors to uniformly orient in space, these efforts must include a component that deals with lidar anisotropic conditions. It was implicit in the design of one of the first laboratory studies of artificial ice cloud backscattering that the means to change the laser viewing angle could be of great importance [8], but many modern lidar systems are restricted to zenith, or near-zenith, pointing directions.

Although I cannot envision a major new lidar application that has not already been tested, several polarization lidar techniques remain to be revisited to examine their potential using improved technologies in the field. These techniques need further exploration in the atmosphere, including Stokes parameters, circular depolarization, and multiwavelength depolarization. In addition, the capabilities of scanning polarization lidars seems particularly promising in the study of the (perhaps) typically anisotropic state of the cloudy atmosphere with respect to the lidar geometry. Lidar depolarization may also prove quite useful in multiparametric relationships to characterize both clouds and aerosols, such as combined  $\delta$ , lidar ratio, and color ratio methods from the ground or Earth orbit.

Finally, the polarization lidar technique offers the potential for monitoring the calibration of Mie and spectroscopic lidars and for correcting multiple-scattering-induced errors that have begun to be explored only fairly recently [51, 52]. This aspect will undoubtedly grow in usage because of the relative ease of adding polarization channels to any lidar system.

## Acknowledgments

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## Lidar and Multiple Scattering

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The various types of lidar applications described in this book differ by the goals pursued and the transceiver configurations designed to meet these goals. However, all lidars rely on the same basic physical process: scattering by discrete scatterers. The common framework for signal analysis and parameter retrieval is almost invariably the single scattering approximation. Under this approximation, the returned lidar signal originates from a single scattering event at an angle determined by the chosen transmitter–receiver geometry but it is attenuated by all preceding and following events. Actually, the extinction caused by multiple scattering is fully taken into account in the single scattering lidar equation. This extinction process is, of course, not what is meant here by multiple scattering. Rather, we mean the part of the multiply scattered radiation that actually ends up being collected by the receiver but that is still considered lost by the single scattering models. In this chapter, we will review and discuss the effects multiple scattering has on the most common applications, describe some calculation methods, and look at proposed ways of correcting for or exploiting these additional contributions.

Multiple scattering is a widespread phenomenon in nature. For instance, it was extensively studied in the field of nuclear physics in which neutron transport plays a major role; see, for example, Davison [1], Case and Zweifel [2], Bell and Glasstone [3], and Williams [4]. The governing equations are basically the same for neutron and optical scattering. However, in lidar applications, the scattering medium is often characterized by a scattering pattern highly peaked in the forward direction, and the transmitters and receivers have generally narrow angular

apertures. For these reasons, the formulation of multiple scattering in lidar, while borrowing a great deal from neutron transport theory, has developed into a specialized field that is worth a separate chapter in a book like this one.

We briefly review in a first section some important lidar single scattering methods that are useful to introduce and understand multiple scattering. Next, we look at how multiple scattering was historically found to affect lidar returns and we present measurement results that illustrate these findings.

We describe in a third section various calculation methods that have been proposed in recent years. Finally, we discuss in the fourth section the corrections that can be applied to the single scattering solutions and we report in Section 3.5 on progress made in the use of multiple scattering as an additional source of retrievable information on particle properties.

Multiple scattering in lidar is a relatively recent research field still in very active evolution. The work described here represents a view of the current state of the art. The scope of this short chapter is necessarily limited and some particular developments may have been missed or left out.

### 3.1 Pertinence of Multiple Scattering

To better recognize the pertinence of multiple scattering in lidar, it is helpful to begin with a brief review of work accomplished in the realm of single scattering. Single scattering solutions are discussed at length elsewhere in this book. We recall here only the basic principles for situations of moderate to high particle densities where multiple scattering effects are likely to be more significant.

The single scattering backscatter lidar equation is presented in Chapter 1. It is simply rewritten here in the form

$$P_{ss}(z) = \frac{K(z)}{z^2} \beta(z) \exp[-2\gamma(z)], \quad (3.1)$$

where  $P_{ss}(z)$  is the single scattering power on the detector from range  $z$ ,  $K(z)$  is the instrument function assumed given,  $\beta(z)$  is the backscattering coefficient,  $\gamma(z) = \int_0^z \alpha(z') dz'$  is the optical depth, and  $\alpha(z)$  is the extinction coefficient. Even for cases in which the molecular contributions to  $\alpha$  and  $\beta$  are negligible, Eq. (3.1) shows that the retrieval of either  $\alpha$  or  $\beta$  constitutes an underspecified problem: one equation for two unknowns.



Rearranging the terms in Eq. (3.1), defining

$$S(z) = P_{ss}(z)z^2/K(z), \quad (3.2)$$

and differentiating with respect to  $z$ , we obtain the following ordinary nonlinear differential equation:

$$\frac{1}{\beta(z)} \frac{d\beta(z)}{dz} - 2\alpha(z) = \frac{1}{S(z)} \frac{dS(z)}{dz}. \quad (3.3)$$

For homogeneous media, we have the additional equation  $d\beta/dz = 0$  which leads, upon substitution in Eq. (3.3), to the simple slope method

$$\alpha(z) = -\frac{1}{2S(z)} \frac{dS(z)}{dz}, \quad (3.4)$$

where  $S(z)$  depends on the measured power  $P_{ss}(z)$  and the known function  $K(z)$ . The intricacies related to determining  $K(z)$  are discussed in other chapters. Note, however, that in the frequent occurrences of constant  $K(z)$  the solution is independent of  $K$ , which means that no calibration is required.

For inhomogeneous media, it is customary to assume a relation between extinction and backscatter of the form

$$\beta(z) = C\alpha^u(z), \quad (3.5)$$

where both  $C$  and  $u$  are constants. While there is little theoretical ground to justify Eq. (3.5) in general, it does not constitute an exceedingly restrictive condition in many practical atmospheric situations [5]. Using Eq. (3.5) in (3.3), we have

$$\frac{u}{\alpha(z)} \frac{d\alpha(z)}{dz} - 2\alpha(z) = \frac{1}{S(z)} \frac{dS(z)}{dz}. \quad (3.6)$$

Note that the constant  $C$  is factored out of Eq. (3.6). Equation (3.6) is a nonlinear ordinary differential equation of elementary structure known as the Bernoulli or homogeneous Riccati equation. The solution is

$$\alpha(z) = \frac{S(z)^{1/u}}{S(z_f)^{1/u}/\alpha(z_f) + \frac{2}{u} \int_z^{z_f} S(z')^{1/u} dz'}, \quad (3.7)$$

where  $z_f$  is the range at which the boundary value  $\alpha(z_f)$  is specified. Note that the integral term in the denominator of Eq. (3.7) is positive

if  $z_f > z$  but negative if  $z_f < z$ . Note also that the solution is again independent of  $K$  if  $K$  is independent of  $z$ . The major difficulty with this solution is that it is unstable in media of moderate to high density unless the boundary value is given at the far end of the measurable lidar return [6] where, however, it is less likely to be known. In addition, the solution (3.7) rests on the validity of Eq. (3.5) which means that the size distribution and composition of the scattering particles must change in a prescribed manner within the medium—the inhomogeneities being solely caused by fluctuations in number density. Further discussions on this solution can be found in Klett [6], Fernald [5], and Bissonnette [7].

Despite the well-defined theoretical basis for solutions (3.4) and (3.7) and their variants, no general application method is possible because the uncertainties on the boundary value and the backscatter-to-extinction relation (3.5) always lead to particular situations. Two groups have proposed expanded measurement techniques that provide additional lidar-derived independent data on  $\alpha$  and  $\beta$  to resolve this problem.

A group at the University of Wisconsin [8–11, cf. also Chapter 5] has developed a high-spectral-resolution lidar transceiver that allows discriminating between Mie and Rayleigh backscattering. The principle is that the Rayleigh backscatter is significantly Doppler broadened by the large thermal velocities of the air molecules whereas the frequency content of the Mie backscatter is nearly unaffected by the slow particle velocities. The two spectra are superposed with the narrow Mie spectrum centered on the broadened Rayleigh spectrum. The separation technique, therefore, requires the use of a high rejection power notch filter centered on the Mie spectrum or laser wavelength. From the known characteristics of the filter, the transmitted and rejected fractions of the aerosol and molecular spectra can be calculated for each detection channel, and the spectra eventually separated. Following the single scattering approximation, we have for the molecular backscatter

$$P_{ss}^m(z) = \frac{K(z)}{z^2} N_m(z) \frac{d\sigma_m(\pi, z)}{d\Omega} \exp[-2\gamma(z)], \quad (3.8)$$

where  $N_m(z)$  is the atmospheric molecular number density and  $d\sigma_m(\pi, z)/d\Omega$  is the differential Rayleigh or molecular scattering cross-section in the backward  $\pi$  direction. From the knowledge of the atmospheric temperature and pressure profiles,  $N_m(z)$  and  $d\sigma_m(\pi, z)/d\Omega$  can be calculated and Eq. (3.8) only depends on  $\alpha(z)$ . Hence, combining Eqs. (3.1) and (3.8) allows the unambiguous determination of both  $\alpha(z)$  and  $\beta(z)$ . The difficulty of the method is technical. In clouds of even

moderate densities, the aerosol backscattering coefficient is much larger than  $N_m d\sigma_m(\pi, z)/d\Omega$ . Since the width of the spectrum from the cloud particles is 4–5 times narrower than the molecular spectrum [9], it turns out that the spectral density at the center can be orders of magnitude greater than on the wings. Hence, to avoid contamination of the molecular signal by the particle backscatter, the rejection power of the filter must be very high. Using an iodine cell, Piironen and Eloranta [11] have achieved a rejection of  $\sim 1:5000$  for a bandwidth of 1.8 pm. A typical bandwidth for the broadened Rayleigh spectrum is 8–10 pm.

Following the same line of thought of deriving additional independent measurements from their lidar, Ansmann et al. [12] proposed to measure, in addition to the elastic backscatter given by Eq. (3.1), the inelastic Raman backscatter off the nitrogen molecules of the atmosphere by use of a filter centered on the nitrogen Raman-shifted laser line. This gives in the single scattering approximation

$$P_{ss}^R(z, \lambda_R) = \frac{K(z)}{z^2} N_{N_2}(z) \frac{d\sigma_m^R(\pi, \lambda_R, z)}{d\Omega} \exp[-\gamma(z, \lambda_0) - \gamma(z, \lambda_R)], \quad (3.9)$$

where  $\lambda_0$  and  $\lambda_R$  are the laser and the  $N_2$ -Raman-shifted wavelengths, respectively,  $N_{N_2}(z)$  is the nitrogen molecular density,  $d\sigma_m^R(\pi, \lambda_R, z)/d\Omega$  is the differential  $N_2$ -Raman cross section in the backward direction, and  $\gamma(z, \lambda_0)$  and  $\gamma(z, \lambda_R)$  are the optical depths at the laser and Raman wavelengths, respectively. The nitrogen number density and the molecular contributions to  $\alpha(z, \lambda_0)$  and  $\alpha(z, \lambda_R)$  are obtainable from the atmospheric temperature and pressure profiles, and the particle extinction coefficients at  $\lambda_0$  and  $\lambda_R$  are related by a simple power law relation [12] that is well justified because  $\lambda_R$  and  $\lambda_0$  are close to one another. That leaves only the aerosol contribution to  $\alpha(z, \lambda_0)$  as the unknown in Eq. (3.9). Thus, by combining the elastic [Eq. (3.1)] and the inelastic [Eq. (3.9)] returns, one can derive the profiles of  $\alpha(z, \lambda_0)$ ,  $\alpha(z, \lambda_R)$  and  $\beta(z, \lambda_0)$  unambiguously and with no further approximations. Here, the wavelengths  $\lambda_R$  and  $\lambda_0$  are sufficiently distant for easy separation, but high measurement precision is a requirement because the Raman signal is orders of magnitude less than the aerosol signal. For comparison, the  $N_2$  Raman cross section is smaller than the atmospheric Rayleigh cross section by a factor of  $\sim 1000$ .

Multiple scattering modifies the picture just described. Figure 3.1 illustrates schematically the scattering events that contribute to the lidar return. The single scattering models take into account only the radiation

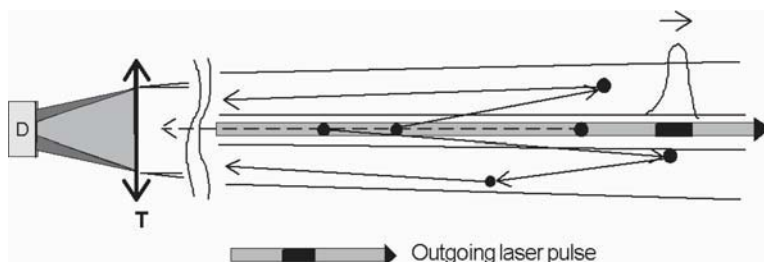


**Fig. 3.1.** Schematic diagram of multiple scattering in the lidar geometry. The thick gray line represents the outgoing laser pulse; the arrows, the scattering events; and the two pairs of thin lines, the limits of two receiver fields of view, a narrow one for near-single-scattering detection, and a wider one for multiple-scattering detection.

scattered once from the outgoing laser pulse back into the receiver, the broken line arrow in Fig. 3.1. However, if the field of view is sufficiently wide and the mean free path between the scattering events sufficiently short for part of the scattered radiation to remain within the field of view, some of it will be re-scattered into the receiver as shown in Fig. 3.1. In most conventional applications, care is taken to keep the field of view as narrow as possible to minimize the multiple scattering contributions but it can never be infinitely small to satisfy the single scattering condition. Therefore, some multiply scattered radiation is always present. In dealing with the conventional solutions (3.4) or (3.7), this extra signal was not of primary concern because of the greater uncertainties associated with the assumption of homogeneity or the specification of the boundary value and the backscatter-to-extinction ratio. With the advent of the high-spectral-resolution and Raman techniques that solve these problems, multiple scattering becomes more pertinent. In moderate to dense media, it can make a significant difference on the calculated solutions.

The obvious multiple scattering effect that is well depicted in Fig. 3.1 is an increase in signal strength. The contributions from the off-axis scattering events are a net addition over the return predicted by the single scattering models. The effect will clearly grow with the field of view and the penetration depth as more and more scattered radiation fills the space seen by the receiver. The measurement geometry is also a driving factor. For the same field of view and medium properties, the amount of diffused radiation within the collecting power of the receiver obviously increases with the distance between the lidar and the medium boundary. A second driving parameter is particle size. It can be seen from the diagram of Fig. 3.1 that the number of light path segments that can be

found within a given receiver field of view increases in inverse proportion to the average angle of the forward scattering events in both the outgoing and return propagation legs. In other words, the amount of collected multiple scattering radiation grows inversely with the angular width of the forward peak of the scattering phase function. For particles of sizes comparable or larger than the lidar wavelength, the width of the peak is inversely proportional to the average particle size. Hence, the strength of the multiply scattered lidar signal, and particularly its rate of increase with field of view, depends on particle size in addition to particle density.

Another effect that is not illustrated in Fig. 3.1 is the incidence multiple scattering has on the polarization state of the lidar signal. It is a well-known fact that the radiation backscattered at exactly  $180^\circ$  by spherical particles conserves the linear polarization of the original laser beam. This property is extensively used to discriminate between particle types as discussed by Sassen in Ref. [13] and in the chapter of this book on polarization. The depolarization ratio  $\Delta$ , defined as the ratio of the scattered field intensities in the perpendicular and parallel directions to the original laser polarization and calculated by the exact Mie theory for a distribution of spherical water droplets [14], is plotted in Fig. 3.2 as a function of the scattering angle near  $180^\circ$ . One clear feature is that  $\Delta$  quickly jumps from 0 to 60% within  $2^\circ$  of the exact backscatter direction. The same calculations show that  $\Delta$  is less than 1% for all forward angles less than  $30^\circ$ . The diagram of Fig. 3.1 indicates that the contributing multiple scattering paths have several forward scatterings at small

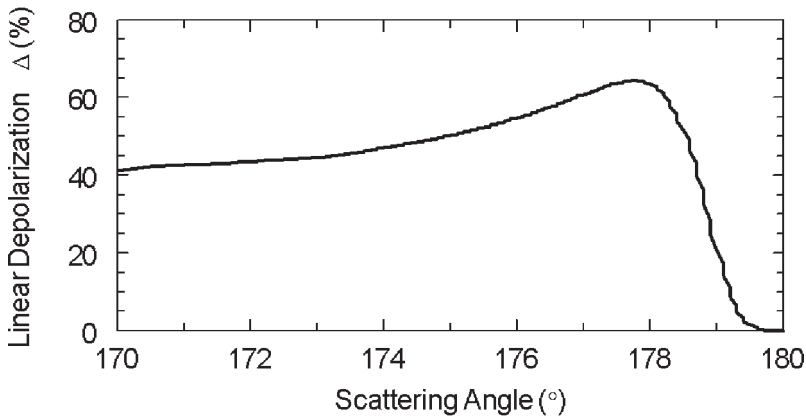


**Fig. 3.2.** Single scattering linear depolarization ratio  $\Delta$  integrated over all azimuth angles calculated for a model C2 Deirmendjian [15] cloud at  $1.06\ \mu\text{m}$  as a function of the scattering angle near the backscattering direction of  $180^\circ$ .

angles and one backscattering at an angle close but not exactly equal to  $180^\circ$ . Given the steep rise of  $\Delta$  near  $180^\circ$ , we can expect depolarization of the multiply scattered lidar returns. Since the calculated  $\Delta$  is negligible at the small forward angles, we further argue that the depolarization arises almost exclusively from the single backscattering near  $180^\circ$ . This is confirmed by the experimental work of Ryan et al. [16, 17] in simulated laboratory water clouds; they found a linear depolarization ratio less than 1% for forward scattered light at optical depths up to 5 [16] but as large as 30% for backscattered light in the same conditions [17]. We expect the degree of depolarization induced by multiple scattering to depend on the same factors as those governing the increase in signal strength.

In summary, multiple scattering in lidar manifests itself as greater signal strength and alteration of polarization state. These effects depend on the measurement geometry, in particular the distance to the scattering medium and the physical penetration depth; on the system parameters, most importantly the receiver field of view; and on the medium properties, i.e., the extinction coefficient, the angular scattering function, and the optical depth. This constitutes a great challenge for modeling but much depends on it. First, the restoration of the solution accuracy of single scattering retrievals in the case of contaminated measurements; and second, the exploitation of the information carried by the multiple scattering contributions. We discuss in the following sections the existing experimental evidence of multiple scattering effects in lidar and the progress made to understand, model and use the data.

### 3.2 Experimental Evidence

Multiple scattering in lidar has long been recognized. One of the first published work that explicitly mentioned multiple scattering are the measurements by Milton et al. [18] of the reflectance of laser-illuminated fair-weather cumulus clouds. Their application was not strictly speaking a lidar experiment because it did not involve ranging but the physics was basically the same. They measured a reflectance of 2–3 times the value derived from single scattering calculations and they attributed the discrepancy to multiple scattering. There followed some discussions [19, 20] centered on the use of a pulsed laser for the experiment but a cw source for the calculations. The controversy had to do with differences in depth of integration between the two configurations. Second-order

theoretical modeling by Anderson and Browell [21] showed that, if the effective pulse length becomes comparable to the scattering mean free path ( $= 1/\alpha$ ), the twice-scattered contribution amounts to more than 20% of the singly scattered return [20]. It is obvious following these discussions that, despite the limitations of the models of the time, second and higher order scattering had to be taken into account to explain the higher-than-expected cloud reflectance. This was later confirmed by Pal et al. [22] in a laboratory-simulated water-droplet cloud.

Platt [23] carried out simultaneous lidar and radiometric measurements on cirrus clouds. He found that the optical depths of the measured cirrus derived by conventional lidar methods was less than the true values. He hypothesized that second- and higher-order scattering processes were responsible for the observed stronger lidar signals. He proposed to redefine the optical depth in the lidar equation (3.1) by inserting a multiplicative factor  $\eta$  to model the reduction of the extinction coefficient as follows:

$$\gamma'(z) = \int_{z_b}^{z_b+h} \eta \alpha(z') dz', \quad (3.10)$$

where  $\alpha$  is the true extinction coefficient,  $z_b$  is the range to cloud base, and  $h$  is the cloud physical thickness. The parameter  $\eta$  is not constant and Platt argued that it should vary between  $\sim 0.5$  and 1. In Ref. [23],  $\eta$  was estimated from the cloud optical thickness determined by radiometric observations and theoretical modeling for the wavelength extrapolation.

The correction factor  $\eta$  is a simple way of representing the premier effect of multiple scattering in lidar, namely, the increase in signal strength. It is still widely used today. One conceptual weakness of the  $\eta$  model is that multiple scattering does not affect only the optical depth  $\gamma$  but also the backscattering coefficient  $\beta$  of Eq. (3.1). There could be situations where the induced drop in the effective backscattering coefficient is greater than the gain caused by the added multiply scattered contributions, which would translate in a value of  $\eta$  greater than unity.

The second aspect of multiple scattering in lidar, i.e., the alteration of the polarization state of the returned signal, was also observed early on. Pal and Carswell [24] designed a lidar system to measure the backscattering of a linearly polarized laser pulse in the parallel and transverse polarization directions simultaneously. They pointed their lidar at water-droplet stratocumulus clouds. They measured a significant perpendicular component that continued to build up for 40–50 m beyond the range where the parallel component reached its peak. Ratioing the two signal

intensities to calculate the linear depolarization ratio  $\delta$ , they found a  $\delta$  that started at a low value of 1–2% at the base of the clouds and increased monotonically with penetration depth to reach values as high as 50% at maximum range. The low depolarization at cloud base indicates that they were probing clouds of spherical water droplets. The subsequent increase in  $\delta$  was rightly attributed to multiple scattering contributions. Indeed, the number of forward scattering events increases and the average angle of backscattering moves away from  $180^\circ$  with rising optical depth; both effects contribute to a greater proportion of depolarized or unpolarized light as can be inferred from Fig. 3.2. But there are other factors affecting  $\delta$ . For example, Pal and Carswell [24] report results from a multilayered cloud deck which show a depolarization ratio that drops suddenly at the transition to a new layer before increasing again. This is explained by the localized dependence of the single scattering return on the coefficient  $\beta$  compared with the more gradual buildup of multiple scattering which is a process of integration. Hence, multiple-scattering-induced depolarization is also a function of cloud structure. Very similar results on cross-polarized lidar returns are also reported by Cohen [25].

The group at York University pursued further their multiple scattering lidar investigations on the basis of depolarization measurements. They proposed the following simple model [26]:

$$P_{\parallel} = P_{\parallel}^s + P_u^m, \quad (3.11)$$

$$P_{\perp} = P_u^m, \quad (3.12)$$

where  $P_{\parallel}$  and  $P_{\perp}$  are the return intensities measured in the parallel and perpendicular directions to the incident laser pulse polarization, the superscripts  $s$  and  $m$  stand for single and multiple scattering, and the subscript  $u$  for unpolarized. The model assumes complete depolarization of the collected multiply scattered radiation. With the model of Eqs. (3.11) and (3.12), they were able to estimate the ratio  $M$  of the total to the single scattering returns, i.e.,

$$M = P_{total}/P_{ss} = (P_{\parallel} + P_{\perp})/(P_{\parallel} - P_{\perp}), \quad (3.13)$$

and the optical depth reduction  $\gamma_m = \gamma - \gamma'$  caused by multiple scattering, i.e.,

$$2\gamma_m(z) = \ln[P_{\parallel}/P_{\parallel}^s] = \ln[P_{\parallel}/(P_{\parallel} - P_{\perp})]. \quad (3.14)$$



They found values of  $M$  of up to 3 and  $\gamma_m$  of up to 0.3–0.4 for penetration depths of 100–120 m in cumulus clouds.

The hypothesis of complete depolarization by multiple scattering made in Eqs. (3.11) and (3.12) was later found by the York group to be incorrect. In Ref. [22], the model was re-defined by assuming that part of the scattered radiation retains the original polarization of the emitted pulse. That polarized component was denoted  $P_{\parallel}^m$  and Eq. (3.11) was rewritten as follows:

$$P_{\parallel} = P_{\parallel}^s + P_{\parallel}^m + P_u^m. \quad (3.15)$$

To be able to isolate  $P_{\parallel}^s$  in this case, they used a field stop to shield the view of the outgoing laser beam. They found that the polarized multiple scattering component  $P_{\parallel}^m$  amounted to  $\sim 40\%$  of the total multiple scattering signal ( $P_{\parallel}^m + P_u^m$ ). It is worth mentioning that the depolarization method described by Eqs. (3.12) and (3.15) only works under the hypothesis that the probed medium is composed of spherical particles.

The York group completed their depolarization analysis by a seminal paper [27] showing that the multiple scattering halo of lidar returns has strongly preferred azimuthal polarization directions. The observed patterns are very sensitive to the size of the scatterers.

Center-blocked field stops and depolarization were also used by Allen and Platt [28] to isolate the multiple scattering component of lidar returns and measure it with greater resolution. They carried out experiments on mixed-phase clouds. Of particular interest is one instance where the depolarization ratio clearly marks the transition between a region of ice crystals and one of spherical droplets. The ratio is shown to drop suddenly at the interface and rise again monotonically by the action of multiple scattering. Our earlier comment on the requirement of purely spherical particles to relate unambiguously depolarization to multiple scattering still holds, but this result shows that depolarization may still be useful in complex situations where the different particle types are not uniformly mixed.

Sassen and Petrilla [29] made measurements of the backscatter and linear depolarization ratio  $\delta$  from marine stratus clouds at different receiver fields of view. They observed a good deal of variability of the in-cloud data. However, their results show that, on average,  $\delta$  increases with penetration depth from a low value of 2.5–4% at the base of the clouds, lower than the subcloud values of 3–6%, in agreement with the data of the York group. Their observed  $\delta$ 's pass through a maximum and then tend to decrease toward the apparent cloud tops. The maximum  $\delta$

depends almost linearly on the field of view measured in units of the matched transmitter/receiver aperture. A typical maximum depolarization ratio for a cloud at 450 m above ground level and a field of view of 3 mrad is 20–30%. In addition, by varying the lidar elevation angle, they found that  $\delta$  does not scale with the vertical or the slant range. This is yet another confirmation that the depolarization induced by multiple scattering is not a local but an integral property that depends on the detailed cloud profile along the lidar path.

Werner et al. [30] studied multiple scattering in lidar as a source of retrievable information on cloud microphysics. In accordance with the observations made in the preceding paragraphs, they identify multiple fields of view, depolarization and pulse stretching as practical means of measuring the multiple scattering contributions. The measurement of the multiply scattered lidar contributions have also been systematically pursued by a lidar group in Canada [31–33]. They have developed multiple-field-of-view (MFOV) depolarization lidars of various designs.

The diagram of a current MFOV lidar design [34] is shown in Fig. 3.3. It incorporates features that allow measurement of the main multiple scattering effects in the small-angle lidar geometry. The receiver field of view is changed at the laser repetition frequency of 100 Hz by rotating a 125-mm diameter aluminized glass disk, shown in an inset in Fig. 3.3, with apertures of different sizes etched at equidistant angular intervals.

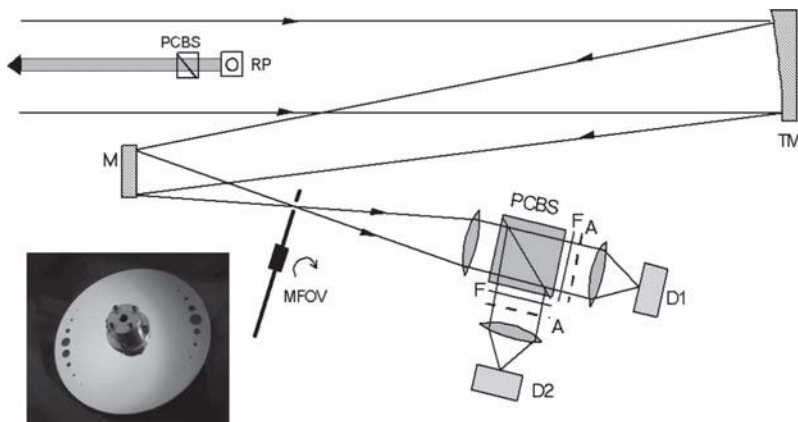


**Fig. 3.3.** Diagram of an existing 1.06- $\mu\text{m}$  multiple-field-of-view lidar. TM: telescope off-axis parabolic mirror; M: plane mirror; MFOV: multiple-field-of-view aperture disk; PCBS: polarizing cube beam splitter; F: narrow-band interference filter; A: attenuator; D1 & D2: silicon avalanche photodiodes; and inset: photograph of field-of-view aperture disk.

The disk is positioned in the image plane of the main telescope mirror. The laser Q-switch is slaved to the disk rotation velocity to ensure that the FOV apertures are in position on the lidar optical axis in synchronization with the laser pulses. The disk has 32 apertures defining 32 FOVs between 0.1 and 12 mrad, full angle. A complete FOV scan takes 32/100 s during which time most clouds can be assumed to remain unchanged. After passage through the FOV aperture, the collected radiation is collimated, separated into parallel and perpendicular polarization components, and focused on 3-mm-diameter Si avalanche photodiodes.

Examples of multiply scattered depolarized returns from a continental cloud deck are plotted in Fig. 3.4 as functions of height above ground level. The lidar wavelength was  $1.06\text{ }\mu\text{m}$ . The returns are truncated where the total signal drops to the smallest power resolved by the detection system and the depolarization ratio where either of the parallel or perpendicular components reaches this limit. This explains why the depolarization curves have shorter stretches than the backscatter curves. Figure 3.4 summarizes very well the experimental evidence of multiple scattering in lidar accumulated over the past 25–30 years and briefly discussed in the preceding paragraphs. We clearly see that the multiple scattering contributions grow with penetration depth from little difference between the different fields of view at the base of the cloud to a

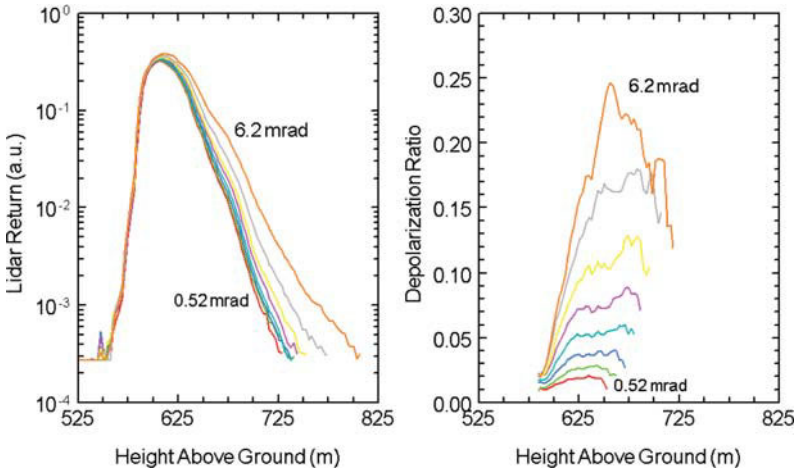


**Fig. 3.4.** Lidar returns (left) and linear depolarization ratios (right) as functions of height above ground for different receiver fields of view. Measurements at a wavelength of  $1.06\text{ }\mu\text{m}$  from a continental stratus cloud deck. Fields of view from bottom to top curves are 0.52, 0.70, 0.96, 1.30, 1.79, 2.44, 3.89, and 6.20 mrad, half angle.

factor of  $\sim 10$  between the largest and smallest fields of view at maximum depth. We note also in Fig. 3.4 that the return at 6.2 mrad penetrates 80 m deeper into the cloud which cannot be explained by pulse stretching for the geometry under which the data were collected. Therefore, there is a significant fading reduction caused by multiple scattering as observed in the early work and modeled by Platt's factor  $\eta$ . Measuring the average slope on the far side of the maximum of the 6.2-mrad and 0.52-mrad curves, we find  $\eta \simeq 0.65$ , i.e., well within the  $0.5 < \eta < 1$  bracket predicted by Platt [23].

The curves of the linear depolarization ratio  $\delta$  of Fig. 3.4 reveal the same basic features as already discussed.  $\delta$  rises from a low value of 1–2% at cloud base to a maximum at a range that depends on the field of view. Except at the large field of view of 6.2 mrad, the maximum is not sharp, the curves mostly level off and no subsequent drop is observed within the measurement precision of the instrument. The maximum value depends on the field of view but is everywhere less than 25%. This is comparable with the findings of Sassen and Petrilla [29] which, however, showed a more pronounced depolarization fall beyond the maximum. Their measurements were for a thin cloud layer in which the maximum penetration, it was argued, coincided with the true cloud top. This, we have seen, is not the case for the experiment reported in Fig. 3.4. Note that the depolarization ratio at 0.52 mrad, which matches the beam divergence, is everywhere less than 2%. Larger values of 5–10% were observed by Sassen and Petrilla [29] for the matched conditions which, however, were equal to 1 mrad in their case. From these findings and observations, we conclude that the linear depolarization ratio is indeed a clear indicator of multiple scattering in the presence of spherical scatterers but, quantitatively,  $\delta$  does not appear to follow simple scaling laws. Modeling depolarization requires that one take into account the complete measurement geometry and the detailed history of the lidar pulse through the inhomogeneities in particle concentration and size. With the added complexity of non-spherical particles, it would seem that the exploitation of depolarization will be more complex than that of the total backscatter dependence on the field of view.

Another important aspect of multiple scattering in lidar that has not been addressed in most measurements reported above is time stretching of the returned pulse. Range is generally calculated assuming that the zigzag multiple scattering paths of Fig. 3.1 have the same length as twice the straight line to the backscattering event. If the receiver field-of-view (FOV) footprint,  $\text{FOV} \cdot z$ , is less than the scattering mean free path equal

to  $1/\alpha$ , there are only a few forward scattering events contributing to the collected return and they occur at angles of the order of the diffraction angle induced by the particles, i.e.,  $\sim \lambda/d_e$ , where  $\lambda$  is the lidar wavelength and  $d_e$  is the effective particle diameter. Under this condition, the path length increase at the  $i$ th scattering is  $\sim i(\lambda^2/d_e^2)/(2\alpha)$ , where  $1/\alpha$  is the “photon” mean free path and  $\sqrt{i}\lambda/d_e$  is the polar angle of its trajectory. Hence, after  $N$  forward scatterings, the total path increase  $\Delta z$  is given by

$$\Delta z \sim \sum_{i=1}^N \frac{i}{2\alpha} \frac{\lambda^2}{d_e^2} \sim \frac{N(N+1)}{4\alpha} \frac{\lambda^2}{d_e^2}. \quad (3.16)$$

Taking  $N$  approximately equal to the optical depth  $\gamma \sim \alpha(z - z_b)$  where  $z_b$  is the range to the boundary of the scattering medium and  $(z - z_b)$  is the penetration depth of the lidar pulse, we find

$$\frac{\Delta z}{z - z_b} \sim \frac{\gamma + 1}{4} \frac{\lambda^2}{d_e^2} \text{ provided that } \text{FOV} < \frac{(z - z_b)}{z} \frac{1}{\gamma}. \quad (3.17)$$

Note that we have not made  $N$  equal to twice the optical depth as would be expected for round-trip propagation because only about one half of the scattering events are small-angle diffraction scatterings. For ground-based applications on low-level clouds with  $(z - z_b) \sim 200$  m,  $z \sim 2$  km, and  $\gamma \sim 3$ , the condition on approximation (3.17) becomes  $\text{FOV} < 35$  mrad, half angle. This condition is almost always satisfied in conventional ground-based lidar systems. Typical lidar wavelengths are of the order of  $1 \mu\text{m}$  and the droplet effective diameter in water clouds is  $\sim 10\text{--}20 \mu\text{m}$ . Hence, the relative path increase  $\Delta z/(z - z_b)$  is less than  $\sim 0.01$  at  $\gamma \sim 3$ . In other words, for ground-based applications, pulse stretching induced by multiple scattering amounts to less than 1% of the penetration depth and it is justified to neglect it. Werner et al. [30] measured  $\Delta z$  through a ground fog over a distance of 150 m. For ground fog,  $d_e \sim 5 \mu\text{m}$  and the approximation (3.17) gives  $\Delta z \sim 6$  m which agrees quite well with the value estimated from their plotted results.

The situation is quite different for long ranges. For example, the epochmaking LITE (Lidar In-space Technology Experiment) experiment [35] was conducted from an orbit altitude of 260 km. Therefore the condition  $\text{FOV} < (z - z_b)/(z\gamma)$  for neglecting pulse stretching was completely violated for dense water clouds, even at the smallest 1.1-mrad field of view of the instrument. In such a geometry, the collected multiple scattering contributions arise from several events at small and large

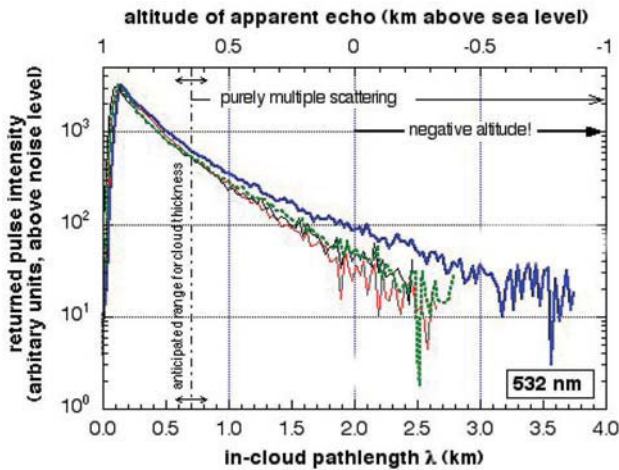


**Fig. 3.5.** Four non-saturated pulses from dense marine stratocumulus clouds observed during NASA's LITE mission [35] of September 1994. Figure created by Anthony Davis (Los Alamos National Laboratory) with LITE data provided by Mark Vaughan (NASA Langley Research Center).

scattering angles, all taking place within the receiver field of view. Sample LITE returns from marine stratus clouds are reproduced in Fig. 3.5. They clearly show a signal above noise not only from a region below the actual cloud base but from negative altitudes, that is, if the range is calculated from the time elapsed after the emission of the laser pulse. The multiply scattered in-cloud path length for these cases is shown in Fig. 3.5 to be greater than 2.5–3 km for an actual thickness of  $\sim 700$  m.

Multiple scattering under receiver field-of-view footprints greater than the scattering mean free path invalidates range resolution based on time of flight. Measurements in such conditions, although different from conventional lidar, are still valid measurements that most certainly contain information on medium properties. New instrument designs and modeling tools are being proposed to exploit this situation. We will discuss those in the following sections.

### 3.3 Modeling

Multiple scattering in lidar is a radiative transfer problem. The radiative transfer theory is defined and developed in several textbooks; see, for

instance, Chandrasekhar [36], Ishimaru [37], and Zege et al. [38]. The fundamental quantity in radiative transfer theory is called the radiance  $I(t, \mathbf{R}, \hat{\mathbf{n}})$ , where  $t$  is the time and  $\mathbf{R}$  and  $\hat{\mathbf{n}}$ , the position and direction vectors, respectively. Bold symbols represent vectors and the superposed hat designates a unit vector. The radiance  $I(t, \mathbf{R}, \hat{\mathbf{n}})$  is the radiant flux per unit solid angle and per unit area normal to  $\hat{\mathbf{n}}$ . To put it differently, the flux  $dP$  through an elementary area  $dS$  normal to  $\hat{\mathbf{n}}_0$  and within the elementary solid angle  $d\Omega$  is given by

$$dP = I(t, \mathbf{R}, \hat{\mathbf{n}}) \hat{\mathbf{n}} \cdot \hat{\mathbf{n}}_0 dS d\Omega. \quad (3.18)$$

$I(t, \mathbf{R}, \hat{\mathbf{n}})$  has the units of  $\text{Wm}^{-2}\text{sr}^{-1}$  and is in general a function of time and six spatial and directional coordinates.

The equation governing the radiance  $I$  is the radiative transfer equation written below in general non-stationary form:

$$\begin{aligned} & \frac{1}{c} \frac{\partial}{\partial t} I(t, \mathbf{R}, \hat{\mathbf{n}}) + \hat{\mathbf{n}} \cdot \nabla_{\mathbf{R}} I(t, \mathbf{R}, \hat{\mathbf{n}}) + \alpha(t, \mathbf{R}) I(t, \mathbf{R}, \hat{\mathbf{n}}) \\ & = \alpha_s(t, \mathbf{R}) \int_{4\pi} I(t, \mathbf{R}, \hat{\mathbf{n}}') p(t, \mathbf{R}; \hat{\mathbf{n}}, \hat{\mathbf{n}}') d\hat{\mathbf{n}}' + Q(t, \mathbf{R}, \hat{\mathbf{n}}), \end{aligned} \quad (3.19)$$

where  $c$  is the speed of light,  $\alpha$  is the extinction coefficient,  $\alpha_s$  is the scattering coefficient,  $p(t, \mathbf{R}; \hat{\mathbf{n}}, \hat{\mathbf{n}}')$  is the scattering phase function, and  $Q(t, \mathbf{R}, \hat{\mathbf{n}})$  is the source/sink term. The phase function is normalized such that

$$\int_{4\pi} p(t, \mathbf{R}; \hat{\mathbf{n}}, \hat{\mathbf{n}}') d\hat{\mathbf{n}}' = 1. \quad (3.20)$$

Equation (3.19) is a heuristic model, it describes the conservation of the radiant flux through an elementary control volume. The particle positions are assumed uncorrelated and their separation wide enough to consider each particle to be in the far field with respect to the radiation scattered by its neighbors. Although diffraction and interference effects are included in the calculation of the scattering and absorption by a single particle, incoherent addition of powers instead of fields is used in constructing  $\alpha_s$  and  $p$  for the particle ensemble and in summing for the radiance  $I$ . In other words, Eq. (3.19) does not follow from the rigorous first principles of Maxwell's equations. However, Ishimaru [37] (Section 14.7) demonstrates that, under certain assumptions, there exists a relationship between  $I$  and the mutual coherence function of the wave field.

Equation (3.19) does not take into account the polarization effects. However, the equation can be readily expanded to include polarization by substituting the Stokes vector for the radiance, and the Mueller matrix for the phase function. The Stokes vector  $\mathbf{I}$  is a four-dimensional vector that describes the polarization state of  $I$  in terms of elementary polarization components. For example, denoting by the subscripts  $\parallel$ ,  $\perp$ ,  $+$  and  $-$  the polarization states of  $I$  measured with linear polarizers oriented in the parallel, perpendicular and tilted at  $\pm 45^\circ$  with respect to the scattering plane, and by  $r$  and  $l$  the states measured through right-hand and left-hand circular polarizers, respectively, we can write the Stokes vector as [39]

$$\mathbf{I} = [(I_{\parallel} + I_{\perp}), (I_{\parallel} - I_{\perp}), (I_{+} - I_{-}), (I_r - I_l)]^T, \quad (3.21)$$

where the superscript T means the transpose operation. The four elements are not all independent but related as follows:

$$(I_{\parallel} + I_{\perp})^2 = (I_{\parallel} - I_{\perp})^2 + (I_{+} - I_{-})^2 + (I_r - I_l)^2. \quad (3.22)$$

The  $4 \times 4$  Mueller matrix is constructed from first principles in the same fashion as the phase function  $p$ . The necessary theoretical tools can be found in Bohren and Huffman [39].

Equation (3.19) is a complex integro-differential equation. No practical general solution exists but numerous approximations have been worked out to handle atmospheric and oceanic transmission problems. Chandrasekhar [36], Ishimaru [37], and Zege et al. [38] give the essential derivation steps for most cases. We will not discuss further these applications in this chapter but concentrate on the lidar.

The lidar geometry is characterized by narrow beams and small receiver fields of view. In addition, the atmospheric or oceanic scattering phase functions at the popular lidar wavelengths are peaked in the forward direction. These particular conditions have led to special solution methods of the radiative transfer equation (3.19). We review in this section the most salient developments for modeling multiple scattering in lidar.

### 3.3.1 Monte Carlo Methods

In view of the serious theoretical and computational difficulties of solving Eq. (3.19), Monte Carlo methods were considered from the beginning as a convenient alternative. The Monte Carlo procedure can be made



to model the same physical processes as the radiative transfer equation but from a statistical approach. The continuous radiance function  $I$  is represented by a very large number of possible distinct trajectories. If one knows the probability for each step in the sequence of events defining a trajectory, or a realization, a possible distinct trajectory can easily be constructed. Then, from many such realizations, averages can be calculated to represent physical quantities, for example, the radiant flux entering a receiver of given position, size and acceptance angle. For the radiative transfer problem, the probabilities are:

1. the probability of scattering or absorption

$$\text{prob}(\text{scatt.}) = \alpha_s / \alpha, \quad (3.23)$$

$$\text{prob}(\text{abs.}) = 1 - \alpha_s / \alpha; \quad (3.24)$$

2. the probability of scattering from direction  $\hat{\mathbf{n}}$  into direction  $\hat{\mathbf{n}}'$  within the elementary solid angle  $d\hat{\mathbf{n}}'$

$$\text{prob}(\hat{\mathbf{n}} \rightarrow \hat{\mathbf{n}}') d\hat{\mathbf{n}}' = p(t, \mathbf{R}; \hat{\mathbf{n}}, \hat{\mathbf{n}}') d\hat{\mathbf{n}}'; \quad (3.25)$$

3. and the probability that the free propagation length from position  $\mathbf{R}$  in the new direction  $\hat{\mathbf{n}}'$  before the next “collision” event is comprised between  $l$  and  $l + dl$

$$\text{prob}(l) dl = \alpha(t, \mathbf{R} + l\hat{\mathbf{n}}') \exp \left[ - \int_0^l \alpha(t, \mathbf{R} + x\hat{\mathbf{n}}') dx \right] dl. \quad (3.26)$$

The needed collision parameters (whether scattering or absorption), photon direction and free path length are calculated by equating the corresponding cumulative probability to a random number uniformly distributed between 0 and 1.

The main advantages of the Monte Carlo approach are that it requires few simplifying approximations, that it allows separation of the contributions by scattering order, and that it can be extended to more complex media with relative ease. For example, to simulate propagation in the presence of various types of scatterers, e.g., molecules, background aerosols, water droplets and ice crystals, it suffices to add to (3.23)–(3.26) the corresponding probabilities for each species.

Radiative transfer Monte Carlo algorithms have been developed first to model solar transmission and backscattering by the atmosphere

and ocean waters. Plass and Kattawar [40–45] were among the first researchers to make systematic use of Monte Carlo simulations in that field.

The implementation of Monte Carlo calculations is rather straightforward but for most radiative transfer problems in the backscatter geometry, techniques of variance reduction are necessary because the angular scattering is generally peaked in the forward direction which makes the probability of a series of events leading to the actual capture of a photon by a backscatter receiver extremely small. This leads to large fluctuations in the calculated averages that can be reduced by increasing the number of trajectories to unrealistic levels or, more practically, by lowering the variance with the help of computational and analytical means.

Variance reduction can be fairly sophisticated. In their work already cited, Plass and Kattawar assigned a statistical weight to each photon, forced collisions within the physical domain of interest so as not to waste calculated trajectories, and then renormalized the photon weight to adjust for the imposed bias. That proved insufficient in lidar simulations because of the very small fields of view of conventional receivers. A significant improvement was achieved by adding analytic calculations to the stochastic Monte Carlo calculations. This method is known as the method of statistical estimation [46]. It is simply hinted in Plass and Kattawar [47] but it is described as a main feature in Kunkel and Weinman [48] and Poole et al. [49, 50]. The method consists in calculating analytically at each collision the probability that the photon would return directly to the receiver without further interactions. This probability is given by

$$\text{prob}(\mathbf{R}_c) = \begin{cases} (-\hat{\mathbf{n}}_r \cdot \hat{\mathbf{n}}_{cr}) \frac{A}{\pi L^2} p(t, \mathbf{R}_c; \hat{\mathbf{n}}_c, \hat{\mathbf{n}}_{cr}) \\ \quad \times \exp \left[ - \int_0^L \alpha(t, \mathbf{R}_r - x \hat{\mathbf{n}}_{cr}) dx \right] & \text{if } (-\hat{\mathbf{n}}_r \cdot \hat{\mathbf{n}}_{cr}) \geq \cos(\Theta), \\ 0 & \text{if } (-\hat{\mathbf{n}}_r \cdot \hat{\mathbf{n}}_{cr}) < \cos(\Theta), \end{cases} \quad (3.27)$$

where  $\mathbf{R}_r$  is the position of the receiver,  $\mathbf{R}_c$  is the position of the scattering or collision event,  $\hat{\mathbf{n}}_r$  is the unit vector normal to the receiver aperture,  $\hat{\mathbf{n}}_{cr}$  is the direction from the collision point to the receiver, i.e.,  $\hat{\mathbf{n}}_{cr} = (\mathbf{R}_r - \mathbf{R}_c)/|\mathbf{R}_r - \mathbf{R}_c|$ ,  $\hat{\mathbf{n}}_c$  is the unit vector giving the direction of the photon prior to the collision,  $A$  is the receiver aperture area,  $L$  is the distance between the receiver and the scattering event given by

$L = |\mathbf{R}_r - \mathbf{R}_c|$ , and  $\Theta$  is the half-angle receiver field of view. The sum of all probabilities for all collision events is used as the backscattered signal instead of the sum of the very few photons which actually would scatter into the receiver. This technique reduces considerably the variance of the calculated signal because all collisions within the receiver field of view contribute. It was validated against Monte Carlo simulations performed without full application of the technique [49] and against measurements in a cell of turbid water [50].

An additional type of a simple but efficient variance reduction method was introduced by Platt [51]. The rationale was to increase the number of photon trajectories in the backward direction. As already discussed, the backscatter probability calculated from (3.25) is very small for most particulate media and lidar wavelengths of practical interest. Hence, there are very few “physical” photons contributing to the signal. To favor more backward trajectories, Platt [51] proposed to create an artificial phase function  $p_a$  by folding the forward half of the true phase function  $p$  into the backward direction as follows:

$$p_a(t, \mathbf{R}; \hat{\mathbf{n}}, \hat{\mathbf{n}}') = \begin{cases} Cp(t, \mathbf{R}; \hat{\mathbf{n}}, \hat{\mathbf{n}}') & \text{for } \hat{\mathbf{n}} \cdot \hat{\mathbf{n}}' \geq 0, \\ Cp(t, \mathbf{R}; \hat{\mathbf{n}}, -\hat{\mathbf{n}}') & \text{for } \hat{\mathbf{n}} \cdot \hat{\mathbf{n}}' < 0, \end{cases} \quad (3.28)$$

where  $C$  is the normalization constant given by  $C^{-1} = 2 \int_{\hat{\mathbf{n}} \cdot \hat{\mathbf{n}}' \geq 0} p(t, \mathbf{R}; \hat{\mathbf{n}}, \hat{\mathbf{n}}') d\hat{\mathbf{n}}'$ . To compensate for the artificially increased number of backward trajectories, the scattered photon is weighted after each collision by the ratio

$$w = p(t, \mathbf{R}; \hat{\mathbf{n}}, \hat{\mathbf{n}}') / p_a(t, \mathbf{R}; \hat{\mathbf{n}}, \hat{\mathbf{n}}'). \quad (3.29)$$

Another method proposed by Brusaglioni et al. [52, 53] consists in defining virtual cloud profiles by adapting the cloud depth or density to each scattering order. The purpose is to optimize the statistics of a given scattering order contribution by adjusting the cloud depth or density (or both) to have on average a number of scattering collisions equal to the scattering order. For example, the virtual extinction coefficient for scattering order  $i$  could be

$$\alpha_i = C_i \alpha \quad \text{with} \quad C_i = 2i \int_0^L \alpha(t, \mathbf{R}_r + x \hat{\mathbf{n}}_r) dx, \quad (3.30)$$

where  $L$  is the physical depth of interest. The total signal is then obtained by summing the contributions of the different scattering orders calculated separately each with a different virtual cloud coefficient  $\alpha_i$ . Of course,

the photon weights have to be renormalized by the ratio of the physical to the virtual probability densities of the free path  $l$ , i.e., by using  $\alpha$  and  $\alpha_i$  in Eq. (3.26), respectively, with  $l$  derived from the virtual cloud cumulative probability. As there are different ways of defining scaling laws of the type given by Eq. (3.30), the reader should consult the original references [52, 53] for details.

Monte Carlo calculations of multiply scattered lidar returns are still slow but they have become accessible to personal computers with the help of the variance reduction techniques such as described above. They are adaptable to very complex problems with no or a minimum of simplifying approximations. No other solution method can match these capabilities and in some cases they are the only tool available. Another important utility of Monte Carlo procedures that follows from their independence on restrictive assumptions is that they can serve as numerical experiments to validate analytical solutions and test inversion methods.

Efficient Monte Carlo codes for simulating lidar returns have been described by Brusaglioni et al. [53], Starkov et al. [54], and Winker and Poole [55]. The three groups compared their calculations for a given ground-based cloud measurement scenario [56]; their results showed very good agreement among themselves. That virtual experiment has led to improvements for handling polarization and inhomogeneities in particle concentration, shape and composition. Results have been presented in numerous conferences and papers, particularly within the MUSCLE (Multiple SCattering Lidar Experiments) group that was responsible for the original intercomparison reported in Ref. [56]. All the important experimental findings on multiple-scattering-induced signal increase and depolarization, and their dependence on measurement geometry, optical depth and medium scattering properties have been demonstrated by Monte Carlo calculations.

One drawback of the Monte Carlo approach is that the solutions are numerical and specific to a given problem. Although there exist a number of scaling relationships as discussed by Brusaglioni et al. [52], one still needs to repeat the calculations for a whole set of values to obtain insight on the influence of a medium or instrument parameter.

### 3.3.2 Stochastic and Phenomenological Methods

It is possible to formulate the problem of multiple scattering in lidar analytically, not in the framework of the radiative transfer equation as it will be the case in the next section, but following the similar premise as

for the Monte Carlo approach. The same notion of random trajectories resulting from successive scattering and absorption events by randomly distributed particles is used. The laws governing the propagation are the same angular scattering and free path distribution functions given by Eqs. (3.25) and (3.26).

Two main groups have used a formal stochastic approach to model the lidar return: Gillespie [57], and Oppel et al. [58] and Starkov et al. [54]. They consider the  $n$ th-order contribution of the power measured at the receiver as the sum over the  $3n$ -dimensional joint probability density of trajectories made up of  $n$  segments, each defined by a free path length and a polar-azimuth heading. The joint probability density is constructed from the elementary functions (3.25) and (3.26). The process is described by Starkov et al. [54] as a generalized Rayleigh's random walk. For example, the end result in Gillespie [57] is a  $3n$ -fold integral over a kernel consisting of the  $n$ -dimensional product of Eqs. (3.25) and (3.26) evaluated at the coordinate points of the integration variables times functions that limit the domain of the integration variables. This expression constitutes an exact multiple scattering lidar equation. The multiple integral can only be computed numerically or by a Monte Carlo method, except for very special cases. For a homogeneous medium, Gillespie [57] was able to transform the integral from  $3n$  to  $(3n - 4)$  dimensions. The final expressions reached by these two groups are not reproduced here because they involve many intermediate definitions that would require more space than available with little additional useful information.

Basically similar expressions have been derived by employing methods of integral calculus. We designate this approach here as phenomenological. In short, the scattered radiation out of an elementary volume is written as the outcome of a scattering event occurring inside this volume on radiation coming from a preceding elementary volume inside which the same type of scattering has taken place. For the  $n$ th-order scattering, there is a cascade of  $n$  such elementary processes. The total contribution is obtained by integrating over all three variables defining each of the  $n$  elementary processes. All integration variables are not independent, they must satisfy a condition on the length of the total path. This constitutes the main practical difficulty of the approach as the integration limits must be chosen from geometrical considerations that increase in complexity with  $n$ . In the stochastic models, the integration limits are over the complete volume; in that case, the constraints took the form of additional kernel functions. Handling the kernel functions probably constitutes a similar difficulty as defining the integration

limits. The stochastic expressions have the advantage of a more general mathematical form. Both methods include time-dependent effects such as pulse stretching since the trajectories are constrained by the time of arrival.

A good example of the phenomenological approach is the model of Cai and Liou [59]. They have derived their solution for the Stokes vector and, therefore, they can calculate any change in polarization state induced by multiple scattering. An adapted version for the received power is reproduced here. According to their model, the power  $P^{(n)}(\tau)$  contributed at the receiver by exactly  $n$  scatterings in a stationary medium characterized by the extinction coefficient  $\alpha(\mathbf{R})$  and the scattering function  $p(\mathbf{R}, \theta)$  with  $\theta = \arccos(\mathbf{n} \cdot \mathbf{n}')$ , and collected at time  $\tau$  following the emission of the laser pulse, is given by

$$\begin{aligned}
 P^{(n)}(\tau) = & P_t \iiint_{V_1} dv_1 \iiint_{V_2} dv_2 \cdots \iiint_{V_n} dv_n \\
 & \times \frac{A}{2\pi R_1^2 [1 - \cos(\psi_m/2)]} \frac{\alpha(\mathbf{R}_n) p(\mathbf{R}_n, \theta_n)}{4\pi R_n^2} \\
 & \times \prod_{i=1}^{n-1} \frac{\alpha_s(\mathbf{R}_i) p(\mathbf{R}_i, \theta_i)}{4\pi |\mathbf{R}_{i+1} - \mathbf{R}_i|^2} \exp \left\{ - \int_0^{R_1} \alpha \left( \frac{\mathbf{R}_1}{R_1} x \right) dx \right. \\
 & - \int_0^{R_n} \alpha \left( \mathbf{R}_n - \frac{\mathbf{R}_n}{R_n} x \right) dx \\
 & \left. - \sum_{i=2}^n \int_0^{|\mathbf{R}_i - \mathbf{R}_{i-1}|} \alpha \left( \mathbf{R}_{i-1} + \frac{\mathbf{R}_i - \mathbf{R}_{i-1}}{|\mathbf{R}_i - \mathbf{R}_{i-1}|} x \right) dx \right\}, \quad (3.31)
 \end{aligned}$$

where  $P_t$  is the laser pulse power evenly distributed within the beam divergence  $\psi_m$ ,  $A$  is the receiver aperture area with diameter assumed much less than any other scale of the problem, and

$$\tau = \frac{1}{c} \left[ R_1 + \sum_{i=2}^n |\mathbf{R}_i - \mathbf{R}_{i-1}| + R_n \right], \quad (3.32)$$

$$\theta_i = \arccos \left( \frac{\mathbf{R}_i \cdot (\mathbf{R}_{i+1} - \mathbf{R}_i)}{R_i |\mathbf{R}_{i+1} - \mathbf{R}_i|} \right) \quad \text{for } i < n, \quad (3.33)$$

$$\theta_n = \arccos \left( - \frac{\mathbf{R}_n \cdot (\mathbf{R}_n - \mathbf{R}_{n-1})}{R_n |\mathbf{R}_n - \mathbf{R}_{n-1}|} \right), \quad (3.34)$$

and  $c$  is the speed of light. In the spherical coordinates  $(R, \phi, \psi)$ , the volume integrals are given by

$$\iiint_{V_i} dv_i \cdots = \int_0^{2\pi} d\phi_i \int_0^{\psi_i^*} d\psi_i \int_{R_i^{**}}^{R_i^*} R_i^2 \sin \psi_i dR_i \cdots \quad (3.35)$$

As already mentioned, the main difficulty is in the determination of the integration limits. Some are straightforward, in particular  $\psi_1^* = \psi_m/2$ , the beam divergence;  $\psi_n^* = \psi_r/2$ , the receiver field of view;  $R_1^* = c\tau/2$ ; and  $R_i^* = H_b(\phi_i, \psi_i)$  for  $i < n$ , the distance to cloud base along the direction defined by  $\mathbf{R}_i$ . The other  $\psi_i^{**}$ 's,  $R_i^{**}$ 's and  $R_n^{**}$  are obtained by finding the bounds of the integration volumes  $V_i$ 's inside which the condition defined by Eq. (3.32) for a fixed  $\tau$  and fixed  $R_j$ 's with  $j < i$  is satisfied. There follows a series of recurrence formulas where the  $R_i^{**}$ 's and  $\psi_i^{**}$ 's for  $i > 2$ ,  $R_n^{**}$  and  $R_n^{**}$  depend on the values derived for the preceding events in the cascade. Hence, the integration limits of Eq. (3.35) can be determined consecutively from  $i = 1$  to  $n$ . The details can be found in Ref. [59].

A recent model by Samoilova [60] including polarization effects and derived from earlier less known Russian work is constructed in the same phenomenological fashion. Another model that also fits this category is that recently proposed by Eloranta [61]. Eloranta considers a simpler situation where all scatterings take place at small forward angles except for one backscattering at an angle close to  $180^\circ$ . Because the angles are small, the photon paths remain close to the lidar axis. As a result, the increase in path length is negligible and the angular integration limits of Eq. (3.35) are much simplified. Eloranta then models the phase function  $p$  for the forward small angles by a Gaussian and assumes uniform backscattering. This enables him to carry out analytically the integrals over the angles  $\phi_i$  and  $\psi_i$ —they become convolutions of Gaussian functions—and the resulting expression is reduced to an  $n$ -dimensional integral over range. The calculation of the solutions, in general, still requires a numerical algorithm but at a much reduced cost. Eloranta goes one step further and shows that, under some special but practical conditions, he can derive simple analytic formulas that provide much physical insight into the dependence of multiple scattering on key medium and instrument parameters. The Gaussian phase function may appear restrictive but the strong diffraction peak of natural cloud and precipitation particles at traditional lidar wavelengths is well represented by

a Gaussian and the resulting solutions are good first approximations for optical depths less than 1–2.

The early second-order scattering work of Liou and Schotland [62], Anderson and Browell [21] and Eloranta [63] can be classified under the phenomenological approach. They followed basically the same derivation steps as described above but for only two scatterings. Therefore, it would seem that the problem formulation was already known 30 years ago or at least 20 years ago for arbitrary scattering order. Progress in applying these tools to correct for or exploit the multiple scattering contributions was slowed by the mathematical complexity of the integrals and the intensive calculations needed to derive numbers. In fact, the stochastic and phenomenological approaches offer little or no computational edge over the Monte Carlo methods. However, the advantage of having formal mathematical expressions is that simplified formulas are possible for limit cases as demonstrated by Eloranta [61]. Even though the accuracy of these simplified formulas may be poor, they are very useful for understanding observations and designing instruments.

### 3.3.3 QSA Approximation—General Theorem

A schematic diagram of multiple scattering in lidar is drawn in Fig. 3.6. The depicted situation is of common occurrence in applications. The instrument wavelength and the medium properties are such that scattering at small angles  $\theta_i$  is predominant. In addition, the footprint of the receiver field of view  $\Theta$  has a diameter less than the mean free path between the scatterings. Under such conditions, the trajectories that contribute most to the received power are made up of small-angle forward scatterings on both the outgoing and return propagation legs and a single backscattering at an angle close to  $180^\circ$ . This regime defines the Quasi-Small-Angle (QSA) approximation of radiative transfer.

Katsev et al. [64] have derived a general theorem that formally simplifies the search for analytic, semianalytic or numerical solutions of the multiple scattering lidar problem in the QSA approximation. In short, they have succeeded in using the Fourier space general solution of the radiative transfer equation in the small-angle approximation limit to define an effective medium with the consequence of transforming the round trip lidar problem into a simpler one-way propagation problem. The mathematics is a little laborious but the result is well worth the effort of going through the main derivation steps.



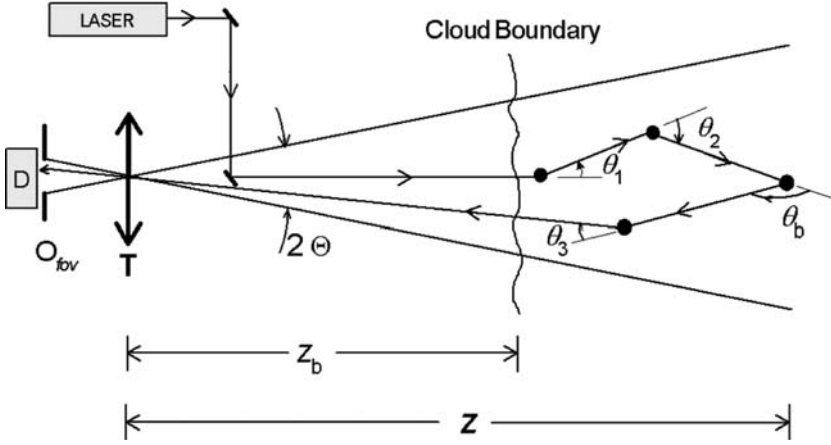


**Fig. 3.6.** Schematic representation of a multiply scattered contribution to lidar return. T: receiver telescope;  $O_{fov}$ : aperture defining the receiver field of view; D: detector;  $z_b$ : range to cloud base;  $z$ : range to backscatter event;  $\theta_i$ : forward scattering angles;  $\theta_b$ : backscattering angle; and  $\Theta$ : half-angle receiver field of view.

In the framework of the QSA approximation and with the help of the diagram of Fig. 3.6, we decompose the lidar radiative transfer problem into the forward propagation of the source radiance on the outgoing path, a diffusion reflection at the backscattering event, the forward propagation of the backscattered radiance on the return path, and the capture by the receiver. All we need to assume at this stage is that there exists a Green's function solution for the radiance propagation. Mathematically, we formulate the problem described above by considering successively

- the source radiance multiply forward scattered to position  $\mathbf{R}$  in the direction  $\hat{\mathbf{n}}$ :

$$I_f(\mathbf{R}, \hat{\mathbf{n}}) = \int d\mathbf{R}_0 \int d\hat{\mathbf{n}}_0 W_{src}(\mathbf{R}_0, \hat{\mathbf{n}}_0) G_o(\mathbf{R}, \hat{\mathbf{n}}; \mathbf{R}_0, \hat{\mathbf{n}}_0), \quad (3.36)$$

where  $W_{src}(\mathbf{R}_0, \hat{\mathbf{n}}_0)$  is the normalized source radiance and  $G_o$  is the Green's function solution of the radiative transfer equation specific to the outgoing problem;

- the radiance singly backscattered at  $\mathbf{R}$  in the direction  $-\hat{\mathbf{n}}_b$ :

$$I_b(\mathbf{R}, -\hat{\mathbf{n}}_b) = \int d\hat{\mathbf{n}} \alpha_{sb}(\mathbf{R}) p_b(\mathbf{R}; -\hat{\mathbf{n}}_b, \hat{\mathbf{n}}) I_f(\mathbf{R}, \hat{\mathbf{n}}), \quad (3.37)$$

where  $\alpha_{sb}$  and  $p_b$  are the scattering coefficient and phase function of the medium constituent responsible for the backscatter;

- the radiance backscattered at  $\mathbf{R}$  that is subsequently forward scattered on the return leg from  $\mathbf{R}$  to a position  $\mathbf{R}''$  and in the direction  $-\hat{\mathbf{n}}''$ :

$$I_r(\mathbf{R}'', -\hat{\mathbf{n}}''; \mathbf{R}) = \int d\hat{\mathbf{n}}_b I_b(\mathbf{R}, -\hat{\mathbf{n}}_b) G_r(\mathbf{R}'', -\hat{\mathbf{n}}''; \mathbf{R}, -\hat{\mathbf{n}}_b), \quad (3.38)$$

where  $G_r$  is the Green's function for the return problem (we distinguish between  $G_o$  and  $G_r$  because the wavelength can be different in both cases as in inelastic Raman applications);

- and the collected power at the receiver originating from the location  $\mathbf{R}$  of the backscattering event:

$$P(\mathbf{R}) = \int d\mathbf{R}'' \int d\hat{\mathbf{n}}'' W_{rec}(\mathbf{R}'', -\hat{\mathbf{n}}'') I_r(\mathbf{R}'', -\hat{\mathbf{n}}''; \mathbf{R}), \quad (3.39)$$

where  $W_{rec}$  is the receiver angular-spatial collection pattern.

Making use of the optical reciprocity principle

$$G(\mathbf{R}'', -\hat{\mathbf{n}}''; \mathbf{R}, -\hat{\mathbf{n}}_b) = G(\mathbf{R}, \hat{\mathbf{n}}_b; \mathbf{R}'', \hat{\mathbf{n}}''), \quad (3.40)$$

defining a “receiver” source

$$W_{src}^{rec}(\mathbf{R}'', \hat{\mathbf{n}}'') = W_{rec}(\mathbf{R}'', -\hat{\mathbf{n}}''), \quad (3.41)$$

combining the expressions (3.36)–(3.39) and collecting the terms in logical groups, we find for the received lidar power from position  $\mathbf{R}$

$$\begin{aligned} P(\mathbf{R}) &= \int d\hat{\mathbf{n}}_b \int d\hat{\mathbf{n}} \alpha_{sb}(\mathbf{R}) p_b(\mathbf{R}; -\hat{\mathbf{n}}_b, \hat{\mathbf{n}}) \\ &\quad \times \int d\mathbf{R}_0 \int d\hat{\mathbf{n}}_0 W_{src}(\mathbf{R}_0, \hat{\mathbf{n}}_0) G_o(\mathbf{R}, \hat{\mathbf{n}}; \mathbf{R}_0, \hat{\mathbf{n}}_0) \\ &\quad \times \int d\mathbf{R}'' \int d\hat{\mathbf{n}}'' W_{src}^{rec}(\mathbf{R}'', \hat{\mathbf{n}}'') G_r(\mathbf{R}, \hat{\mathbf{n}}_b; \mathbf{R}'', \hat{\mathbf{n}}''). \end{aligned} \quad (3.42)$$

Finally, if we define the source and receiver–source radiances

$$I_{src}(\mathbf{R}, \hat{\mathbf{n}}) = \int d\mathbf{R}_0 \int d\hat{\mathbf{n}}_0 W_{src}(\mathbf{R}_0, \hat{\mathbf{n}}_0) G_o(\mathbf{R}, \hat{\mathbf{n}}; \mathbf{R}_0, \hat{\mathbf{n}}_0), \quad (3.43)$$

$$I_{src}^{rec}(\mathbf{R}, \hat{\mathbf{n}}) = \int d\mathbf{R}_0 \int d\hat{\mathbf{n}}_0 W_{src}^{rec}(\mathbf{R}_0, \hat{\mathbf{n}}_0) G_r(\mathbf{R}, \hat{\mathbf{n}}; \mathbf{R}_0, \hat{\mathbf{n}}_0), \quad (3.44)$$

as the multiply scattered radiances at position  $\mathbf{R}$  originating from the true source  $W_{src}$  and the “receiver” source  $W_{src}^{rec}$ , respectively, we end up with

$$P(\mathbf{R}) = \int d\hat{\mathbf{n}}_b \int d\hat{\mathbf{n}} \alpha_{sb}(\mathbf{R}) p_b(\mathbf{R}; -\hat{\mathbf{n}}_b, \hat{\mathbf{n}}) I_{src}(\mathbf{R}, \hat{\mathbf{n}}) I_{src}^{rec}(\mathbf{R}, \hat{\mathbf{n}}_b). \quad (3.45)$$

Equation (3.45) reduces the lidar problem into two conceptually simpler propagation problems, defined by Eqs. (3.43) and (3.44), connected by one backscattering event. This results from the QSA description of the lidar multiple scattering problem and the reciprocity principle of optical propagation given by Eq. (3.40).

Next, we turn to the solutions for  $I_{src}(\mathbf{R}, \hat{\mathbf{n}})$  and  $I_{src}^{rec}(\mathbf{R}, \hat{\mathbf{n}})$ . In accordance with the QSA approach, the forward propagation problem satisfies the conditions of the small-angle approximation (SAA). One important simplification is that the medium can be considered stratified. This is justified by the smallness of the scattering angles,  $\theta_i$  in Fig. 3.6, and the narrow angular width of both the source and receiver functions  $W_{src}$  and  $W_{src}^{rec}$ . Thus, the  $\mathbf{R} = z\hat{\mathbf{k}} + \mathbf{r}$  dependence of the medium properties is reduced to a  $z$  dependence and the direction vector  $\hat{\mathbf{n}}$  is approximated by  $\hat{\mathbf{n}} \simeq \hat{\mathbf{k}} + \mathbf{n}_\perp$ , where  $\hat{\mathbf{k}}$  is the unit vector along the  $z$  axis chosen to coincide with the lidar axis,  $\mathbf{r}$  is the component of the position vector normal to  $\hat{\mathbf{k}}$ , and  $|\mathbf{n}_\perp| \ll 1$ . Since  $|\mathbf{n}_\perp| \ll 1$ ,  $\mathbf{n}_\perp$  is the vector scattering angle, i.e.,  $\mathbf{n}_\perp \simeq \theta[\hat{\mathbf{i}} \cos \phi + \hat{\mathbf{j}} \sin \phi]$  where  $\theta$  and  $\phi$  are the polar and azimuth components of the radiance angular variable, respectively. The above approximations essentially amount to making  $\sin \theta \simeq \theta$  and  $\cos \theta \simeq 1$ , and to neglecting path increase caused by beam spreading. Moreover, because  $\mathbf{n}_\perp$  is small, the phase function for randomly oriented particles depends only on the difference  $|\hat{\mathbf{n}}_\perp - \hat{\mathbf{n}}'_\perp|$  rather than on  $\hat{\mathbf{n}}_\perp$  and  $\hat{\mathbf{n}}'_\perp$  independently. Under these conditions, the stationary version of the radiative transfer equation (3.19) with no source term becomes, in the forward direction,

$$\begin{aligned} \frac{\partial}{\partial z} I(z, \mathbf{r}, \mathbf{n}_\perp) + \mathbf{n}_\perp \cdot \nabla_r I(z, \mathbf{r}, \mathbf{n}_\perp) + \alpha(z) I(z, \mathbf{r}, \mathbf{n}_\perp) \\ = \alpha_s(z) \int_{-\infty}^{\infty} I(z, \mathbf{r}, \mathbf{n}'_\perp) p(z; |\mathbf{n}_\perp - \mathbf{n}'_\perp|) d\mathbf{n}'_\perp, \end{aligned} \quad (3.46)$$

where the integration limits on  $\mathbf{n}'_\perp$  were extended to  $\pm\infty$  because, within the SAA,  $p(z; |\mathbf{n}_\perp - \mathbf{n}'_\perp|)/p(z; 0)$  becomes negligibly small with increasing  $|\hat{\mathbf{n}}_\perp - \hat{\mathbf{n}}'_\perp|$ .

Both  $I_{src}(\mathbf{R}, \hat{\mathbf{n}})$  and  $I_{src}^{rec}(\mathbf{R}, \hat{\mathbf{n}})$  are solutions of Eq. (3.46) with their respective phase functions  $p_o$  and  $p_r$ . Because the integral term in Eq. (3.46) is a convolution, it is feasible to obtain a formal solution in the Fourier space. Following Ishimaru [37] (Section 13.2) or Zege et al. [38] (Section 4.4.3), the solutions are

$$\tilde{I}_{src}(z, \mathbf{q}, \mathbf{p}) = \tilde{W}_{src}(\mathbf{q}, \mathbf{p} + \mathbf{q}z) \tilde{G}_o(z, \mathbf{q}, \mathbf{p}), \quad (3.47)$$

$$\tilde{I}_{src}^{rec}(z, \mathbf{q}, \mathbf{p}) = \tilde{W}_{src}^{rec}(\mathbf{q}, \mathbf{p} + \mathbf{q}z) \tilde{G}_r(z, \mathbf{q}, \mathbf{p}), \quad (3.48)$$

where the Fourier transforms are defined by

$$\tilde{I}(z, \mathbf{q}, \mathbf{p}) = \int_{-\infty}^{\infty} d\mathbf{r} \int_{-\infty}^{\infty} d\mathbf{n}_{\perp} I(z, \mathbf{r}, \mathbf{n}_{\perp}) e^{(-i\mathbf{q}\cdot\mathbf{r} - i\mathbf{p}\cdot\mathbf{n}_{\perp})}, \quad (3.49)$$

$$\tilde{W}_{src}(\mathbf{q}, \mathbf{p}) = \int_{-\infty}^{\infty} d\mathbf{r} \int_{-\infty}^{\infty} d\mathbf{n}_{\perp} W_{src}(0, \mathbf{r}, \mathbf{n}_{\perp}) e^{(-i\mathbf{q}\cdot\mathbf{r} - i\mathbf{p}\cdot\mathbf{n}_{\perp})}, \quad (3.50)$$

$$\tilde{p}(z, \mathbf{p}) = \int_{-\infty}^{\infty} d\mathbf{n}_{\perp} p(z, \mathbf{n}_{\perp}) e^{-i\mathbf{p}\cdot\mathbf{n}_{\perp}}, \quad (3.51)$$

with  $\tilde{W}_{src}$  standing for both  $\tilde{W}_{src}$  and  $\tilde{W}_{src}^{rec}$ , and the Green's function solution  $\tilde{G}$  of the Fourier transformed Eq. (3.46) is

$$\begin{aligned} \tilde{G}_{o,r}(z, \mathbf{q}, \mathbf{p}) = \exp \left\{ - \int_0^z [\alpha_{o,r}(z - \xi) - \alpha_{s(o,r)}(z - \xi) \right. \\ \left. \times \tilde{p}_{o,r}(z - \xi, \mathbf{p} + \mathbf{q}\xi)] d\xi \right\}. \end{aligned} \quad (3.52)$$

The function  $W_{src}(0, \mathbf{r}, \mathbf{n}_{\perp})$  of Eq. (3.50) represents the boundary value for both sources  $W_{src}$  and  $W_{src}^{rec}$ , and the subscript  $(o, r)$  in Eq. (3.52) stands for either  $o$  or  $r$ .

Rewriting the expression (3.45) for the lidar signal  $P(\mathbf{R})$  in the SAA variables, we have

$$\begin{aligned} P(z, \mathbf{r}) = \int d\mathbf{n}_{\perp b} \int d\mathbf{n}_{\perp} \alpha_{sb}(z) p_b(z; -\hat{\mathbf{k}} - \mathbf{n}_{\perp b}, \hat{\mathbf{k}} - \mathbf{n}_{\perp}) \\ \times I_{src}(z, \mathbf{r}, \mathbf{n}_{\perp}) I_{src}^{rec}(z, \mathbf{r}, \mathbf{n}_{\perp b}). \end{aligned} \quad (3.53)$$

Since the angles  $\mathbf{n}_{\perp}$  and  $\mathbf{n}_{\perp b}$  are small, the phase function  $p_b$  depends on the difference  $|\mathbf{n}_{\perp b} - \mathbf{n}_{\perp}|$  only and we can write

$$p_b(z; -\hat{\mathbf{k}} - \mathbf{n}_{\perp b}, \hat{\mathbf{k}} - \mathbf{n}_{\perp}) = p_b(z; \pi - |\mathbf{n}_{\perp b} - \mathbf{n}_{\perp}|). \quad (3.54)$$

Furthermore, we neglect in the SAA the path length increase caused by the forward scattering zigzags and we equate the range  $z$  to the time  $\tau$  following the emission of the laser pulse, i.e.,  $z = c\tau/2$ . Hence, the lidar return measured at time  $\tau$ , or from range  $z$ , is approximated by integrating Eq. (3.53) over  $\mathbf{r}$  in the plane  $z$ . Changing the angular integration variables from  $\mathbf{n}_{\perp b}$  and  $\mathbf{n}_{\perp}$  to  $\mathbf{n}_{\perp d} = \mathbf{n}_{\perp b} - \mathbf{n}_{\perp}$  and  $\mathbf{n}_{\perp}$ , we therefore have for the lidar signal from range  $z$

$$P(z) = \int d\mathbf{r} \int_{-\infty}^{\infty} d\mathbf{n}_{\perp d} \int_{-\infty}^{\infty} d\mathbf{n}_{\perp} \alpha_{sb}(z) p_b(z; \pi - |\mathbf{n}_{\perp d}|) I_{src}(z, \mathbf{r}, \mathbf{n}_{\perp}) \times I_{src}^{rec}(z, \mathbf{r}, \mathbf{n}_{\perp} + \mathbf{n}_{\perp d}). \quad (3.55)$$

We have extended in Eq. (3.55) the angular integration limits to  $\pm\infty$  because the function  $I_{src}$  and  $I_{src}^{rec}$  rapidly become negligible with increasing  $\mathbf{n}_{\perp d}$  and  $\mathbf{n}_{\perp}$ . Finally, for convenience, we rewrite Eq. (3.55) as follows:

$$P(z) = \int_{-\infty}^{\infty} d\mathbf{n}_{\perp d} \alpha_{sb}(z) p_b(z; \pi - |\mathbf{n}_{\perp d}|) H(z, \mathbf{n}_{\perp d}), \quad (3.56)$$

with

$$H(z, \mathbf{n}_{\perp d}) = \int d\mathbf{r} \int_{-\infty}^{\infty} d\mathbf{n}_{\perp} I_{src}(z, \mathbf{r}, \mathbf{n}_{\perp}) I_{src}^{rec}(z, \mathbf{r}, \mathbf{n}_{\perp} + \mathbf{n}_{\perp d}). \quad (3.57)$$

Because the solutions  $\tilde{I}_{src}$  and  $\tilde{I}_{src}^{rec}$  are derived in Fourier space, we Fourier transform Eq. (3.57) and apply the Parseval equality [65]. We thus obtain for symmetric functions of  $\mathbf{p}$  and  $\mathbf{n}_{\perp}$

$$\tilde{H}(z, \mathbf{p}) = \frac{1}{(2\pi)^2} \int d\mathbf{q} \tilde{I}_{src}^*(z, \mathbf{q}, \mathbf{p}) \tilde{I}_{src}^{rec}(z, \mathbf{q}, \mathbf{p}), \quad (3.58)$$

where the  $*$  indicates the complex conjugate of the function. The Parseval equality states that the integral in physical space of the product of one function by the complex conjugate of another is equal to the integral in Fourier space of the product of their corresponding transforms. Substituting the solutions (3.47) and (3.48) for  $\tilde{I}_{src}$  and  $\tilde{I}_{src}^{rec}$  in Eq. (3.58), we have

$$\tilde{H}(z, \mathbf{p}) = \frac{1}{(2\pi)^2} \int d\mathbf{q} \tilde{W}_{src}^*(\mathbf{q}, \mathbf{p} + \mathbf{q}z) \tilde{W}_{src}^{rec}(\mathbf{q}, \mathbf{p} + \mathbf{q}z), \times \tilde{G}_o^*(z, \mathbf{q}, \mathbf{p}) \tilde{G}_r(z, \mathbf{q}, \mathbf{p}). \quad (3.59)$$

Using the general solution (3.52), we define an effective Green's function

$$\begin{aligned}\tilde{G}^e(z, \mathbf{q}, \mathbf{p}) &= \tilde{G}_o^*(z, \mathbf{q}, \mathbf{p}) \tilde{G}_r(z, \mathbf{q}, \mathbf{p}) \\ &= \exp \left\{ - \int_0^z [\alpha^e(z - \xi) - \alpha_s^e(z - \xi) \tilde{p}^e(z - \xi, \mathbf{p} + \mathbf{q}\xi)] d\xi \right\},\end{aligned}\quad (3.60)$$

where

$$\alpha^e = \alpha_o + \alpha_r \quad (3.61)$$

$$\alpha_s^e = \alpha_{so} + \alpha_{sr} \quad (3.62)$$

$$p^e = \frac{\alpha_{so} p_o + \alpha_{sr} p_r}{\alpha_{so} + \alpha_{sr}} \quad (3.63)$$

are the properties of the effective medium. In constructing the expression (3.63) for the equivalent phase function  $p^e$ , we have assumed that  $p_o(z; \mathbf{n}_\perp)$  is an even function of  $\mathbf{n}_\perp$  so that  $\tilde{p}_o^* = \tilde{p}_o$ . This is always the case for random orientation of scatterers. Note that for elastic backscattering, i.e., identical properties for the outgoing and return propagation legs, the effective medium has twice the extinction and scattering coefficients but the same phase function as the true medium. Regrouping the terms inside the integral of Eq. (3.59), we define

$$\tilde{I}^{*e}(z, \mathbf{q}, \mathbf{p}) = \tilde{W}_{src}^*(\mathbf{q}, \mathbf{p} + \mathbf{q}z) \tilde{G}^e(z, \mathbf{q}, \mathbf{p}), \quad (3.64)$$

$$\tilde{W}(z, \mathbf{q}, \mathbf{p}) = \tilde{W}_{src}^{rec}(\mathbf{q}, \mathbf{p} + \mathbf{q}z), \quad (3.65)$$

where the equivalent radiance  $\tilde{I}^e$  and receiver function  $\tilde{W}$  will be given proper interpretations below.  $\tilde{H}(z, \mathbf{p})$  of Eq. (3.59) can thus be rewritten

$$\tilde{H}(z, \mathbf{p}) = \frac{1}{(2\pi)^2} \int d\mathbf{q} \tilde{W}(z, \mathbf{q}, \mathbf{p}) \tilde{I}^{e*}(z, \mathbf{q}, \mathbf{p}). \quad (3.66)$$

Now that we have obtained  $\tilde{H}$  in terms of the radiance solution for the effective medium defined by Eqs. (3.61)–(3.63), we inverse Fourier transform Eq. (3.66) to return to the physical space. Applying the Parseval equality to the result, we find for symmetric functions of  $\mathbf{p}$  and  $\mathbf{n}_\perp$

$$H(z, \mathbf{n}_{\perp d}) = \int d\mathbf{r} \int_{-\infty}^{\infty} d\mathbf{n}_\perp W(z, \mathbf{r}, \mathbf{n}_{\perp d} + \mathbf{n}_\perp) I^e(z, \mathbf{r}, \mathbf{n}_\perp). \quad (3.67)$$

Substituting Eqs. (3.67) for  $H(z, \mathbf{n}_{\perp d})$  back into Eq. (3.56), we obtain the following expression for the lidar signal from range  $z$ :

$$P(z) = \int d\mathbf{r} \int_{-\infty}^{\infty} d\mathbf{n}_{\perp d} \int_{-\infty}^{\infty} d\mathbf{n}_{\perp} \alpha_{sb}(z) p_b(z; \pi - |\mathbf{n}_{\perp d}|) \\ \times W(z, \mathbf{r}, \mathbf{n}_{\perp d} + \mathbf{n}_{\perp}) I^e(z, \mathbf{r}, \mathbf{n}_{\perp}). \quad (3.68)$$

From the comparison of Eqs. (3.64) and (3.65) with the formal Fourier space solution, Eq. (3.47) or (3.48), of the SAA radiative transfer equation, the following interpretations for  $I^e$  and  $W$  become obvious:

- $I^e(z, \mathbf{r}, \mathbf{n}_{\perp})$  is the source radiance forward propagated to position  $(z, \mathbf{r})$  from the true source function  $W_{src}(0, \mathbf{r}, \mathbf{n}_{\perp})$  in the effective medium defined by Eqs. (3.61)–(3.63),  $I^e(z, \mathbf{r}, \mathbf{n}_{\perp})$  is called the equivalent or effective radiance; and
- $W(z, \mathbf{r}, \mathbf{n}_{\perp})$  is the actual receiver function  $W_{rec}(z, \mathbf{r}, \mathbf{n}_{\perp})$  since  $\tilde{W}(z, \mathbf{q}, \mathbf{p})$  of Eq. (3.65) is the vacuum ( $\tilde{G} = 1$ ) forward-propagation Fourier solution of the “receiver” source  $W_{src}^{rec}$  defined in Eq. (3.41) in terms of the true receiver pattern.

Equation (3.68) is the main result of this section. It implies that the calculation of multiply scattered lidar returns can be accomplished in the QSA regime by substituting the real medium by a fictitious medium of effective properties given by Eqs. (3.61)–(3.63) in the outgoing path, of same angular backscattering function as the actual medium, and of zero extinction and scattering in the return path. Hence, the round trip lidar problem is replaced by the simpler problem of solving the forward propagation of a radiance beam in a properly specified effective medium. The return propagation is trivial as it takes place in vacuum. In other words, all forward scattering events of Fig. 3.6 are modeled to occur on the outgoing propagation leg with reduced free path lengths because  $\alpha_s^e$  is nearly twice greater than the individual  $\alpha_{so}$  and  $\alpha_{sr}$ . This is illustrated in Fig. 3.7 where no attempt was made to draw an exactly equivalent trajectory because the equivalence exists on the average only. Individual trajectories such as depicted in Fig. 3.7 are virtual representations. Finally, it should be noted that Eq. (3.68) allows for inelastic backscattering;  $\alpha_o, \alpha_r, \alpha_{so}, \alpha_{sr}, \alpha_{sb}, p_o, p_r$ , and  $p_b$  can all be different.

The most common receivers have a pupil area  $\Sigma_{rec}$  of dimension much less than the field-of-view footprint at working ranges and a uniform angular profile delimited by a field of view  $\Theta$  (half angle). With these

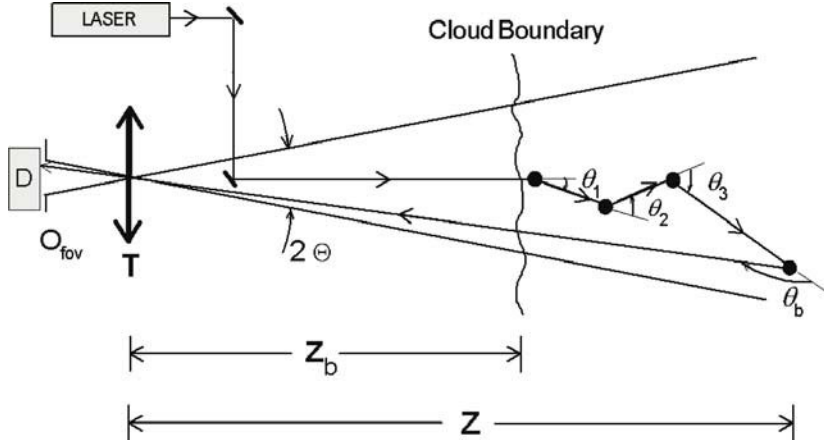


**Fig. 3.7.** Same as in Fig. 3.6 except that the multiple scatterings are drawn in the equivalent effective medium defined by Eqs. (3.61–3.63).

specifications, the function  $W_{rec}$  for  $r/z \ll 1$  has the trivial form

$$W_{rec}(z, \mathbf{r}, \mathbf{n}_\perp) = \begin{cases} (\Sigma_{rec}/z^2)\delta(n_\perp - r/z) & \text{if } r \leq \Theta z, \\ 0 & \text{otherwise.} \end{cases} \quad (3.69)$$

Using Eq. (3.69) in (3.68), assuming axisymmetry and performing the integral over the  $\delta$  function, we obtain the following practical expression for the lidar return from range  $z$  and within field of view  $\Theta$ :

$$P(z, \Theta) = (2\pi)^2 \frac{\Sigma_{rec}}{z^2} \int_0^{\Theta z} r dr \int_0^\infty n_\perp dn_\perp \alpha_{sb}(z) \times p_b(z, \pi - |r/z - n_\perp|) I^e(z, \mathbf{r}, n_\perp). \quad (3.70)$$

Katsev et al. [64] propose a formulation slightly different from Eq. (3.68). It is somewhat less intuitive but more efficient computationally in cases of simple source and receiver profiles. They define an equivalent source profile by convoluting the true source and receiver functions as follows:

$$\tilde{\mathcal{W}}^e(\mathbf{q}, \mathbf{p} + \mathbf{q}z) = \tilde{W}_{src}^*(\mathbf{q}, \mathbf{p} + \mathbf{q}z) \tilde{W}_{src}^{rec}(\mathbf{q}, \mathbf{p} + \mathbf{q}z). \quad (3.71)$$

Then, the equivalent radiance is derived for the equivalent source and not for the real source as it was done in Eq. (3.64), i.e.,

$$\tilde{\mathcal{I}}^e(z, \mathbf{q}, \mathbf{p}) = \tilde{\mathcal{W}}^e(\mathbf{q}, \mathbf{p} + \mathbf{q}z) \tilde{G}^e(z, \mathbf{q}, \mathbf{p}). \quad (3.72)$$



The effective medium, however, is the same as before, i.e., Eqs. (3.61)–(3.63). Making use of Eqs. (3.47), (3.48), (3.60), (3.71) and (3.72) in Eq. (3.58) yields

$$\tilde{H}(z, \mathbf{p}) = \frac{1}{(2\pi)^2} \int d\mathbf{q} \tilde{\mathcal{I}}^e(z, \mathbf{q}, \mathbf{p}). \quad (3.73)$$

The inverse transform is thus

$$H(z, \mathbf{n}_{\perp d}) = \mathcal{I}^e(z, \mathbf{r} = 0, \mathbf{n}_{\perp d}), \quad (3.74)$$

and Eq. (3.56) for the return signal  $P(z)$  becomes

$$P(z) = \int_{-\infty}^{\infty} d\mathbf{n}_{\perp d} \alpha_{sb}(z) p_b(z; \pi - |\mathbf{n}_{\perp d}|) \mathcal{I}^e(z, \mathbf{r} = 0, \mathbf{n}_{\perp d}), \quad (3.75)$$

or in axisymmetric conditions

$$P(z) = 2\pi \int_0^{\infty} dn_{\perp d} \alpha_{sb}(z) p_b(z; \pi - n_{\perp d}) \mathcal{I}^e(z, \mathbf{r} = 0, n_{\perp d}). \quad (3.76)$$

The receiver function, that appears explicitly in Eq. (3.68) or through the field of view  $\Theta$  and angle  $r/z$  in Eq. (3.70), is embedded here in the effective radiance. For example, a new  $\mathcal{I}^e(z, \mathbf{r} = 0, \mathbf{n}_{\perp d})$  has to be calculated for each different field of view but there is no integration over  $r$  compared with Eq. (3.70). In the physical space, the effective source profile is calculated from the inverse Fourier transform of Eq. (3.71), which leads to a convolution. The effective medium is the same for both  $\mathcal{I}^e$  and  $I^e$ .

The solution given by Eq. (3.68) or (3.75) is general. For particular applications, one needs to derive expressions for the Fourier transforms of the phase functions and of the true or effective source profile, and to substitute the results in Eq. (3.64) or (3.72). Then the functions  $\tilde{I}^e$  and  $\tilde{\mathcal{I}}^e$  have to be inverse Fourier transformed to calculate the physical effective radiances of Eq. (3.68) or (3.75). To this day, there are no known exact methods for performing these tasks because of the complexity of the phase functions. Note, however, that there is no requirement to use the Fourier approach to solve for  $I^e$  or  $\mathcal{I}^e$ ; any valid solution can be used in Eq. (3.68) or (3.75).

The general result expressed by Eq. (3.68) or (3.75) constitutes a significant simplification of the problem of multiple scattering in lidar where the QSA approximation is justified. In particular, the convolution of the Green's function in Eq. (3.42) that involves a fourfold integration

on the scattering angles has been eliminated. The problem becomes one of forward propagation only. Compared with the stochastic and phenomenological methods of the preceding section, the number of nested integrations is reduced considerably for scattering orders of 7–8 needed at fields of view and optical depths of practical measurements.

### 3.3.4 QSA Approximation—A Neumann Series Solution

One early application of the QSA approach to calculate multiply scattered lidar signals is the model of Weinman [66] and Shipley [67]. Their basic definition of the problem is the same as illustrated in Fig. 3.6 and used throughout the derivations of Subsection 3.3.3, namely: small-angle forward scatterings in both the outgoing and return paths and a single backscattering event at an angle close to 180°. Weinman also assumes, without demonstration, that the backscattered radiance captured within the narrow fields of view of conventional lidar receivers propagates basically in the same manner as the outgoing radiance. This working hypothesis, made twenty years earlier, has nearly the same effect on calculations as the effective-medium theorem of Katsev et al. [64] discussed above. Another aspect of special interest to this chapter is that Weinman makes use of the Neumann series that transforms the integro-differential radiative transfer equation into a set of purely differential equations.

The Neumann series is the simple series

$$I = \sum_{n=1}^{\infty} I_n. \quad (3.77)$$

Equation (3.77) is substituted for  $I$  in the SAA radiative transfer equation (3.46) and the individual  $I_n$ 's are chosen to satisfy

$$\begin{aligned} \frac{\partial}{\partial z} I_n + \mathbf{n}_{\perp} \cdot \nabla_r I_n + \alpha I_n \\ = (1 - \delta_{n1}) \alpha_s \int_{-\infty}^{\infty} I_{n-1}(z, \mathbf{r}, \mathbf{n}'_{\perp}) p(z; \mathbf{n}_{\perp} - \mathbf{n}'_{\perp}) d\mathbf{n}'_{\perp}, \end{aligned} \quad (3.78)$$

with  $\delta_{n1}$  defined by

$$\delta_{ij} = \begin{cases} 1 & \text{if } i = j, \\ 0 & \text{if } i \neq j. \end{cases} \quad (3.79)$$

The radiance  $I_n$  is interpreted as the contribution from the  $n$ th scattering order. The integral term in Eq. (3.78) becomes a known function of the solutions obtained at the lower orders. The series begins with  $I_1$  which is the trivial solution for the unscattered radiance. Hence, the Neumann series solution method removes the difficulty of dealing with an integro-differential equation and it has the advantage of distinguishing between the scattering orders. The algorithm proposed by Weinman [66] is a good example of its application.

Weinman further simplifies Eq. (3.78) by relating the variable  $\mathbf{r}$  to the scattering angle  $\theta = \arccos(\mathbf{n}_\perp \cdot \mathbf{n}'_\perp)$  instead of treating  $\mathbf{r}$  and  $\mathbf{n}_\perp$  as fully independent variables. More precisely, he sets  $(\mathbf{r} \cdot \mathbf{r}) = \ell^2 \theta^2$ , where  $\ell$  is the scattering free path. Equation (3.78) is next reduced to a one-dimensional ordinary differential equation by Fourier transforming with respect to the transverse spatial coordinate. Finally, to make the resulting solutions integrable analytically over the angular and transverse coordinates, Weinman models the phase function as a finite sum of Gaussian functions. The final expression for  $I_n$  has the form of  $(n - 1)$  nested integrations over the axial distance  $z$  which need to be performed numerically. The integration steps  $\Delta z$  must be made small enough to ensure negligible probability of a second scattering within  $\Delta z$ . The complete expression is not reproduced here as many parameter, variable and function definitions are required for a satisfactory interpretation; the interested reader can find the details in Ref. [66].

The computation time increases rapidly with increasing scattering order. However, for experiments conducted at fields of view chosen as small as possible to minimize multiple scattering, the number of necessary orders can be kept as low as 4–5 and the computation load remains acceptable up to optical penetration depths of  $\sim 3$  encountered with real systems in real situations. Therefore, the method has definite practical merit. It was successfully used by Wandinger [68] to estimate multiple scattering effects in Raman and high-spectral-resolution lidars. Wandinger also showed in the same work that the method produces results in excellent agreement with the MUSCLE group Monte Carlo simulations reported in [56].

### 3.3.5 QSA Approximation—Analytic Solutions

A great deal of theoretical work was carried out in Russia on analytic and semianalytic solutions of the SAA radiative transfer equation.

A representative example of this effort applied to the lidar problem is reported by Zege et al. [69].

A first method begins with expanding in Taylor series, with respect to the angular variable  $\mathbf{n}'_{\perp}$ , the radiance  $I(z, \mathbf{r}, \mathbf{n}'_{\perp})$  in the integral of Eq. (3.46). Assuming that  $I$  is symmetric in  $\mathbf{n}'_{\perp}$  and carrying out the expansion up to the quadratic term, we have

$$I(z, \mathbf{r}, \mathbf{n}'_{\perp}) = I(z, \mathbf{r}, \mathbf{n}_{\perp}) + \frac{|\mathbf{n}_{\perp} - \mathbf{n}'_{\perp}|^2}{2} \nabla_{n_{\perp}}^2 I(z, \mathbf{r}, \mathbf{n}_{\perp}) + \dots \quad (3.80)$$

For validity, the second term in Eq. (3.80) must be considerably smaller than the leading term. Substituting Eq. (3.80) into (3.46), we find

$$\begin{aligned} \frac{\partial}{\partial z} I(z, \mathbf{r}, \mathbf{n}_{\perp}) + \mathbf{n}_{\perp} \cdot \nabla_r I(z, \mathbf{r}, \mathbf{n}_{\perp}) + [\alpha(z) - \alpha_s(z)] I(z, \mathbf{r}, \mathbf{n}_{\perp}) \\ = \frac{\alpha_s(z) \beta^2(z)}{2} \nabla_{n_{\perp}}^2 I(z, \mathbf{r}, \mathbf{n}_{\perp}), \end{aligned} \quad (3.81)$$

where

$$\begin{aligned} \beta^2(z) &= 2\pi \int_0^{\infty} |\mathbf{n}_{\perp} - \mathbf{n}'_{\perp}|^3 p(z; |\mathbf{n}_{\perp} - \mathbf{n}'_{\perp}|) d|\mathbf{n}_{\perp} - \mathbf{n}'_{\perp}|, \\ &\simeq 2(1 - g), \end{aligned} \quad (3.82)$$

and  $g$  is the asymmetry factor defined by  $g = 2\pi \int_{-1}^1 \cos \theta p(z, \theta) d \cos \theta$ . To be consistent with the second-order Taylor expansion, the  $\cos |\mathbf{n}_{\perp}|$  that was approximated as unity in front of the  $z$  derivative in Eq. (3.46) must be reinstated in Eq. (3.81). We thus obtain using Eq. (3.82)

$$\begin{aligned} \left[ 1 - \frac{|\mathbf{n}_{\perp}|^2}{2} \right] \frac{\partial}{\partial z} I(z, \mathbf{r}, \mathbf{n}_{\perp}) + \mathbf{n}_{\perp} \cdot \nabla_r I(z, \mathbf{r}, \mathbf{n}_{\perp}) + [\alpha(z) - \alpha_s(z)] \\ \times I(z, \mathbf{r}, \mathbf{n}_{\perp}) = \alpha_s(z)(1 - g) \nabla_{n_{\perp}}^2 I(z, \mathbf{r}, \mathbf{n}_{\perp}). \end{aligned} \quad (3.83)$$

The angular scattering properties are all embedded in the asymmetry factor. Further details on this approximation, called the small-angle diffusion approximation, can be found in Zege et al. [38, 69, 70].

Zege et al. derive in Ref. [69] an analytic solution of Eq. (3.83) by assuming a Gaussian radiance profile of the form

$$I(z, \mathbf{r}, \mathbf{n}_{\perp}) = A(z) \exp[-|\mathbf{n}_{\perp} - B(z)\mathbf{r}|^2 / C(z)]. \quad (3.84)$$

Expressions for  $A$ ,  $B$  and  $C$  are found by solving the ordinary differential equations obtained by substitution of Eq. (3.84) for  $I$  in

Eq. (3.83). Details and more references can be found in Zege et al. [38] (Sections 4.5.1–4.5.3).

The solution (3.84) was worked out by Zege et al. [69] for an effective source profile and scattering medium defined by Eqs. (3.71) and (3.61)–(3.63), respectively. Substituting the resulting expression  $\mathcal{I}^e(z, \mathbf{r} = 0, \mathbf{n}_\perp)$  in Eq. (3.75) and assuming a multicomponent Gaussian model for the backscatter function  $p_b$ , they obtained an analytic expression for the multiply scattered lidar return. They compared their predictions with Monte Carlo calculations. All the features of the Monte Carlo simulations are well reproduced but the analytic solutions underestimate the multiple scattering contributions by a factor of  $\sim 2$ . Zege et al. trace the origin of this bias to the assumed Gaussian profile of Eq. (3.84) that smooths too much the radiance near  $\mathbf{r} = 0$ , particularly for narrow beams and peaked scattering phase functions. To verify this, they looked at the multiply forward scattered radiance from sources of increasing divergence. They found that the bias indeed decreases rapidly with the divergence. The discrepancies are also less for lidar returns at very large receiver fields of view.

Analytic solutions such as Eq. (3.84) have the obvious advantages of providing instantaneous numbers and useful asymptotic formulas. However, in view of the important underestimation discussed in the preceding paragraph, Zege et al. explored the option of computing  $\mathcal{I}^e(z, \mathbf{r} = 0, \mathbf{n}_\perp)$  by inverse Fourier transforming numerically the solutions obtained through Eqs. (3.72), (3.71) and (3.60). The difficulty in this case is to design an efficient multidimensional integration algorithm which amounts to choosing the optimal grid in the Fourier  $(\mathbf{q}, \mathbf{p})$ -space. The task is somewhat facilitated by the analytic solution (3.84) which provides information on the relevant scales. In the case of Ref. [69], the choice was further aided by splitting the forward peak of the phase function into two components. This semianalytic solution method gives results in excellent agreement with the Monte Carlo simulations of Ref. [56]. There is a non-negligible computational load but it is less than for the phenomenological methods reviewed in this chapter. Furthermore, estimations of the contributions by scattering order can be obtained by expanding in Taylor series the exponential term in Eq. (3.60). Tam and Zardecki [71] developed such a solution in analytic form for a source of Gaussian-angular and  $\delta$ -spatial profiles propagating in a uniform particulate medium having a Gaussian phase function.

The analytic and semianalytic solutions derived by Zege et al. [69] for the one-way propagation of the effective radiance  $I^e$  or  $\mathcal{I}^e$  coupled

with the general lidar Eq. (3.68) or (3.75) are an important step forward in modeling multiple scattering in lidar. They not only constitute efficient computational tools but the general theoretical framework on which they rest provides solid rigorous grounds for the advancement of the inverse problem.

Tam [72] has worked out a two-stream model of the QSA radiative transfer equation to calculate lidar returns. The forward-stream radiance satisfies the radiative transfer Eq. (3.46) except for the phase function that is truncated to the forward hemisphere only. Tam solves the forward problem in Fourier space by expanding the phase function term of Eq. (3.52) as in his earlier work of Ref. [71]. He then simplifies the backward-stream equation making the usual QSA approximations of a peaked forward phase function and a single backscattering but he still needs to solve a three-dimensional partial differential equation with a complicated source term. In view of the effective medium theorem of Subsection 3.3.3, this tedious task would no longer be necessary. This is one good example of the benefit that follows from the effective medium theorem.

### 3.3.6 QSA Approximation—A Semiempirical Solution

Two- and four-flux models have been used extensively in radiative transfer problems. In general, applications have been limited to plane-parallel geometries. Following the work of Tam, Bissonnette [73] proposed a two-flux or two-stream model for narrow beams but with an engineering approach to make computations easier to handle arbitrary media. The radiance  $I$  is split into an unscattered  $I_u$  and a scattered  $I_s$  component. Both components are transformed into irradiances by integrating over the forward and backward hemispheres as follows:

$$\begin{aligned} U_u &= \int_{(2\pi)^+} d\mathbf{n}_\perp I_u(z, \mathbf{r}, \mathbf{n}_\perp) \quad \text{and} \\ U_s^\pm &= \int_{(2\pi)^\pm} d\mathbf{n}_\perp I_s(z, \mathbf{r}, \mathbf{n}_\perp). \end{aligned} \quad (3.85)$$

For simplicity, we assume an infinitely narrow source beam and we have for  $U_u$

$$U_u(z, r) = I_0(z, r) \exp[-\gamma(z)], \quad (3.86)$$

where  $I_0$  is the source beam irradiance and  $\gamma$  is the optical depth. The QSA radiative transfer equation (3.46) is also integrated over the forward

and backward hemispheres. The resulting equations contain the flux functions

$$\mathbf{F}_s^\pm = \int_{(2\pi)^\pm} \mathbf{n}_\perp I_s(z, \mathbf{r}, \mathbf{n}_\perp) d\mathbf{n}_\perp. \quad (3.87)$$

$\mathbf{F}_s^\pm$  are the radiation fluxes in directions transverse to the beam axis. To have a close system of equations,  $\mathbf{F}_s^\pm$  must be related to  $U_s^\pm$ . Here, the model makes the empirical assumption that the lateral fluxes  $\mathbf{F}_s^\pm$  result from a diffusion process defined by

$$\mathbf{F}_s^\pm = -D^\pm(z) \nabla_r U_s^\pm(z, \mathbf{r}). \quad (3.88)$$

The constitutive diffusion relation (3.88) and coefficients  $D^\pm$  are not formally derived from the radiative transfer equation. Instead, by analogy with turbulent transport processes, it is postulated that  $D^\pm$  are proportional to the product of the mean free path between the scattering events that give rise to the lateral flux or transport of  $U_s^\pm$  times the strength of this random “microscopic motion.” Empirical expressions for  $D^\pm$  are derived from this postulate in Ref. [73]. With the help of the defining relation (3.88), a closed set of two differential equations is obtained for  $U_s^\pm$ . Because of the effective medium theorem of Subsection 3.3.3, it now suffices in QSA lidar applications to solve for forward propagation alone. We therefore reproduce here only the forward flux equation,

$$\frac{\partial}{\partial z} U_s^+ - D^+ \nabla_r^2 U_s^+ + (\alpha - \alpha_s^+) U_s^+ = \alpha_s^+ I_0(z, r) \exp[-\gamma(z)], \quad (3.89)$$

in which we have dropped the  $\alpha_s^- U_s^-$  term in accordance with the QSA assumption of a single backscattering and where  $\alpha_s^\pm = \alpha_s \int_{(2\pi)^\pm} d\mathbf{n}_\perp p(z, \mathbf{n}_\perp)$ . Equation (3.89) has a general solution that involves a twofold nested integration over  $z$ . For use in the lidar equation (3.68) or (3.70), we need to reinstate the angular dependence that was integrated in the definition of  $U_s^+$ . This is done in Ref. [73] in an *ad hoc* fashion. Comparisons with laboratory measurements [74, 75] for both transmission and backscattering show good agreement. On the other hand, the MUSCLE intercomparisons of calculated multiply scattered lidar returns [56] indicate that the simultaneous solutions of Eq. (3.89) and its return-stream counterpart give a resulting lidar signal that falls at the limit of the data spread.

The model just outlined incorporates a good deal of empiricism and lacks the mathematical rigor of the preceding solution methods. However, it has the main advantage of making the calculations affordable for

arbitrary media stratified perpendicularly to the lidar axis. The simplification to forward propagation only resulting from Eq. (3.70) improves the accuracy of the lidar solution and further reduces the required computational effort by more than one half. Short computation times are important for retrieval applications where the search for an optimal solution often involves iterations or repeated calculations.

### 3.3.7 Diffusion Limit

The diffusion limit in multiple scattering lidar has not been very much explored in the past because it essentially means that the photon origin is lost and with it the intrinsic ranging property of traditional lidars. However, it has been suggested in recent studies [76, 77] that the off-axis returns can provide key bulk properties of dense diffusing clouds. Time and space simply take on different meanings that can be exploited with the diffusion theory.

We follow here the work of Davis et al. [77]. Since the off-axis measurements of interest have to do with a cloud region large compared with the dimension of the source beam, the latter is modeled by delta functions of time, space and angular spread. We therefore seek a solution of the homogeneous nonstationary radiative transfer equation (3.19) as a response to an impulse, which defines the Green's function. The initial and boundary conditions are

$$G(t, z, \mathbf{r}, \hat{\mathbf{n}}; 0, 0, \mathbf{0}, \hat{\mathbf{k}}) = \begin{cases} \delta(t)\delta(\mathbf{r})\delta(\hat{\mathbf{n}} - \hat{\mathbf{k}}) & \text{for } t > 0; z = 0; \hat{\mathbf{k}} \cdot \hat{\mathbf{n}} \geq 0, \\ 0 & \text{for } t > 0; z = \Delta_z; \hat{\mathbf{k}} \cdot \hat{\mathbf{n}} \leq 0, \\ 0 & \text{for } t = 0; \mathbf{r} \in \mathbb{R}^2; 0 \leq z \leq \Delta_z, \end{cases} \quad (3.90)$$

where  $z = 0$  is the base of the cloud and  $z = \Delta_z$  is the cloud thickness. In the following, we will drop the source coordinates in the Green's function notation since they are constant throughout.

What we want to do here is derive, in the diffusion limit, a solution for  $G$  that satisfies the initial and boundary conditions (3.90). According to Ishimaru [37] (Section 9.1), one main characteristic of the diffusion limit is that  $G$  can be expanded as follows:

$$G(t, z, \mathbf{r}, \hat{\mathbf{n}}) \simeq \frac{1}{4\pi} [J(t, z, \mathbf{r}) + 3\hat{\mathbf{n}} \cdot \mathbf{F}(t, z, \mathbf{r})], \quad (3.91)$$



where  $J$  and  $\mathbf{F}$  are the radiant intensity and flux vector, respectively,

$$J(t, z, \mathbf{r}) = \int G(t, z, \mathbf{r}, \hat{\mathbf{n}}) d\hat{\mathbf{n}}, \quad (3.92)$$

$$\mathbf{F}(t, z, \mathbf{r}) = \int \hat{\mathbf{n}} G(t, z, \mathbf{r}, \hat{\mathbf{n}}) d\hat{\mathbf{n}}. \quad (3.93)$$

Equation (3.91) is the expansion of  $G(t, z, \mathbf{r}, \hat{\mathbf{n}})$  about the condition of uniform angular distribution given by  $G = J/4\pi$ , and hence the second term must be sufficiently smaller than the first, i.e.,  $|\mathbf{F}| \ll J$ . From here on, we will assume that this condition is satisfied and use the equal sign in Eq. (3.91). The constitutive diffusion hypothesis assumed by Davis et al. [77] is

$$\mathbf{F} = -\frac{1}{3}\ell_t(z, \mathbf{r})\nabla_R J(t, z, \mathbf{r}), \quad (3.94)$$

where  $\ell_t$  is the photon transport mean free path given by

$$\ell_t = [\alpha - \alpha_s g]^{-1}, \quad (3.95)$$

and  $g$  is the asymmetry factor. Substituting Eq. (3.94) for  $\mathbf{F}$  in Eq. (3.91), we have

$$G(t, z, \mathbf{r}, \hat{\mathbf{n}}) = \frac{1}{4\pi} [1 - \ell_t(z, \mathbf{r})\hat{\mathbf{n}} \cdot \nabla_R] J(t, z, \mathbf{r}). \quad (3.96)$$

Therefore, the diffusion model of radiative transfer amounts to solving for the radiant intensity  $J$  instead of the radiance. Integrating the radiative transfer equation (3.19) over  $\hat{\mathbf{n}}$ , making the source term  $Q = 0$ , and using the constitutive relation (3.94), we obtain the following equation for  $J$ :

$$\frac{\partial}{\partial t} J - \nabla_R \cdot D \nabla_R J + c(\alpha - \alpha_s) J = 0, \quad (3.97)$$

where  $D$  is called the diffusion coefficient. It is given by

$$D = \frac{c\ell_t}{3} = \frac{c}{3(\alpha - \alpha_s g)}. \quad (3.98)$$

Equation (3.97) is the diffusion equation. In our search for a solution, we will assume in the following that the medium is homogeneous, in other words, that  $D$ ,  $\alpha$ ,  $\alpha_s$  and  $g$  are constants. As it is customary in mathematical physics, we transform the linear partial

differential equation (3.97) into an ordinary differential equation by use of a Fourier–Laplace transform, i.e.,

$$\tilde{J}(s, z, \mathbf{q}) = \iiint \exp[-st + i\mathbf{q} \cdot \mathbf{r}] J(t, z, \mathbf{r}) dt d\mathbf{r}. \quad (3.99)$$

Equation (3.97) thus becomes the ordinary differential equation

$$\frac{d^2}{dz^2} \tilde{J} - \tilde{J}/L^2 = 0 \quad \text{with} \quad L^{-2} = q^2 + s/D + c(\alpha - \alpha_s)/D. \quad (3.100)$$

The Fourier–Laplace transformation of the initial and boundary conditions (3.90) gives with the help of Eq. (3.96) and the assumption of axisymmetry with respect to the  $z$  axis the following mixed boundary conditions for  $\tilde{J}(s, z, \mathbf{q})$ :

$$\begin{aligned} \frac{1}{2} \left[ 1 - \frac{\ell_t}{2} \frac{d}{dz} \right] \tilde{J}(s, z, \mathbf{q}) &= 1 \quad \text{at } z = 0, \\ \frac{1}{2} \left[ 1 + \frac{\ell_t}{2} \frac{d}{dz} \right] \tilde{J}(s, z, \mathbf{q}) &= 0 \quad \text{at } z = \Delta_z. \end{aligned} \quad (3.101)$$

The two-point boundary value problem of Eqs. (3.100) and (3.101) is easily solved. However, the final expression for  $\tilde{J}(s, z, \mathbf{q})$  is quite involved and the inverse transformation back into physical space is not possible. Fortunately, the main observables can be expressed in terms of  $\tilde{J}(s, z, \mathbf{q})$ .

In off-axis lidar applications, the accessible quantity is the reflected flux at  $180^\circ$  from the base of the cloud defined as follows by Davis et al. [77]:

$$G_R(t, \mathbf{r}) = \int_{(2\pi)^-} |\hat{\mathbf{k}} \cdot \hat{\mathbf{n}}| G(t, 0, \mathbf{r}, \hat{\mathbf{n}}) d\hat{\mathbf{n}}. \quad (3.102)$$

Using Eq. (3.96) in (3.102), we obtain the following equation relating  $G_R$  to  $J$ :

$$G_R(t, \mathbf{r}) = \frac{1}{4} \left[ 1 + \frac{2}{3} \ell_t \frac{\partial}{\partial z} \right] J(t, z, \mathbf{r}) \quad \text{at } z = 0. \quad (3.103)$$

Applying the Fourier–Laplace transformation to Eq. (3.103), we find

$$\tilde{G}_R(s, \mathbf{q}) = \frac{1}{4} \left[ 1 + \frac{2}{3} \ell_t \frac{d}{dz} \right] \tilde{J}(s, z, \mathbf{q}) \quad \text{at } z = 0. \quad (3.104)$$

The boundary conditions (3.101) and the defining relations (3.103) and (3.104) are mathematically exact but they assume that the diffusion constitutive relations (3.91) and (3.94) remain applicable down to and including the transition boundary at  $z = 0$ . Obviously, the true radiance at  $z = 0$  is not diffuse for all angles  $\hat{\mathbf{n}}$ . Since Eqs. (3.101), (3.103) and (3.104) contain the radiant intensity evaluated at  $z = 0$ , they cannot be expected to model with full accuracy the true reflected flux. Consequently, researchers [2, 77] have chosen slightly different boundary conditions than Eq. (3.101) because the collimated model of Eq. (3.90) is not compatible with the diffusion limit, and by extension they have also modified the numerical constants of Eqs. (3.103) and (3.104). The proposed alternate boundary conditions and relation between  $\tilde{G}_R$  and  $\tilde{J}$  are [77]

$$\begin{aligned} \frac{1}{2} \left[ 1 - \chi \ell_t \frac{d}{dz} \right] \tilde{J}(s, z, \mathbf{q}) &= 1 \quad \text{at } z = 0, \\ \frac{1}{2} \left[ 1 + \chi \ell_t \frac{d}{dz} \right] \tilde{J}(s, z, \mathbf{q}) &= 0 \quad \text{at } z = \Delta_z, \end{aligned} \quad (3.105)$$

$$\tilde{G}_R(s, \mathbf{q}) = \frac{1}{2} \left[ 1 + \chi \ell_t \frac{d}{dz} \right] \tilde{J}(s, z, \mathbf{q}) \quad \text{at } z = 0, \quad (3.106)$$

where  $\chi$  is an adjustable or free numerical factor of  $O(1)$  to be determined a posteriori, for example by comparisons with Monte Carlo simulations.

From the definition of the Fourier–Laplace transform, Eq. (3.99), we have the following relations between  $\tilde{G}_R$  and a few observables:

$$\tilde{G}_R(0, \mathbf{0}) = \iiint G_R(t, \mathbf{r}) dt d\mathbf{r} = R, \quad (3.107)$$

$$\left| \frac{\partial^u}{\partial s^u} \tilde{G}_R(s, \mathbf{0}) \right|_{s=0} = \iiint t^u G_R(t, \mathbf{r}) dt d\mathbf{r} = \langle t^u \rangle R, \quad (3.108)$$

$$\left| \nabla_q \cdot \nabla_q \tilde{G}_R(0, \mathbf{q}) \right|_{\mathbf{q}=0} = \iiint r^2 G_R(t, \mathbf{r}) dt d\mathbf{r} = \langle r^2 \rangle R, \quad (3.109)$$

where  $R$  is the space-time average cloud reflection,  $c\langle t \rangle$  is the mean photon pathlength,  $c^2\langle t^2 \rangle$  is the mean-square photon pathlength, and  $\langle r^2 \rangle$  is the mean-square horizontal transport length. Therefore, the measurable quantities  $R$ ,  $\langle t \rangle$ ,  $\langle t^2 \rangle$  and  $\langle r^2 \rangle$  can be written through Eqs. (3.106)–(3.109) in terms of the Fourier–Laplace solution  $\tilde{J}(s, z, \mathbf{q})$  of the diffuse intensity. The latter can be easily obtained in analytic form for a homogeneous medium of sufficient density to justify the diffusion limit

approximation by solving the ordinary differential equation (3.100) with the boundary conditions (3.105).

Davis et al. [77] and Love et al. [78] give the following asymptotic expressions for the observables derived under  $\alpha_s = \alpha$  or an albedo  $\omega = \alpha_s/\alpha$  of unity:

$$R = \frac{\Delta_z}{2\chi\ell_t + \Delta_z} = \frac{(1-g)\gamma}{2\chi + (1-g)\gamma}, \quad (3.110)$$

$$c\langle t \rangle = 2\chi\Delta_z + \text{corr. term}, \quad (3.111)$$

$$c^2\langle t^2 \rangle = \frac{4\chi}{5}\Delta_z^2(1-g)\gamma + \text{corr. term}, \quad (3.112)$$

$$\langle r^2 \rangle = \frac{8\chi}{3} \frac{\Delta_z^2}{(1-g)\gamma} + \text{corr. term}, \quad (3.113)$$

where  $\gamma = \alpha\Delta_z$  is the single scattering cloud optical depth, and “corr. term” stands for a term of lesser magnitude than the shown leading term. The leading terms (without precise numerical constants) in Eqs. (3.110)–(3.113) can be derived from heuristic scaling arguments [76]. Although they vanish for large  $\gamma$ , the correction terms are not negligible at commonly observed values of  $\gamma$ , say, 5–50.

From any two of the four relations (3.110)–(3.113), the cloud bulk parameters  $\Delta_z$  and  $\gamma$  can in principle be retrieved from the reflected off-axis halo surrounding the laser beam measured with space-time resolution. These two cloud parameters are the most variable by far; for liquid stratiform clouds, one can confidently set  $g \approx 0.85$  [79] and  $\chi \approx 2/3$  [2]. Off-beam lidar inversions are briefly discussed in Subsection 3.5.3.

### 3.3.8 Summary

The direct problem of calculating multiply scattered lidar returns is now well understood and has been satisfactorily solved by a variety of techniques. The Monte Carlo methods have reached a high degree of maturity. They have the definite advantage of minimizing the number of necessary simplifying approximations and, for this reason, they provide easy virtual experiments for testing other models and retrieval algorithms. Furthermore, except for the demand on computational resources, there is no conceptual difficulty to set up Monte Carlo simulations for problems with complex instrumentation specifications, special measurement geometry and highly structured scattering properties. One disadvantage of Monte Carlo simulations is that the problems are treated one at a

time; solution trends can only be studied through large numbers of runs. The analytic and semianalytic models provide useful assistance in such applications. Despite the simplifying assumptions and the limited precision and range, analytic expressions can readily exhibit many aspects of a problem. In particular, the effective medium theorem of Subsection 3.3.3 is a noteworthy achievement. It casts in rigorous mathematical terms a situation that was hypothesized in some form or other in many previous models.

The models reviewed in this section are representative of the state of the art. They were built on several other contributions that are not all referenced here, especially the early work in the former Soviet Union. The reader should consult Refs. [38, 64, 69, 70] for a more complete bibliography on the latter.

### 3.4 Accounting for Multiple Scattering

The first obvious application of the multiple scattering models is estimating errors on the parameters retrieved with single scattering algorithms and working out corrections. As already mentioned in Section 3.2, Platt [23] introduced the parameter  $\eta$ , Eq. (3.10), to take into account the reduction in the extinction coefficient caused by multiple forward scatterings. He used  $\eta$  to define the effective extinction and backscatter-to-extinction ratio,  $\alpha_e = \eta\alpha$  and  $k_e = \beta/\alpha_e = k/\eta$ , respectively, and indicated [80] that the outputs of single scattering retrievals are actually the effective  $\alpha_e$  and  $k_e$  instead of the true—meaning single scattering—extinction coefficient  $\alpha$  and backscatter-to-extinction ratio  $k$ . If  $\eta$  were a constant independent of range, it would be a simple matter to calculate its value for whole classes of problems and the corrections would be straightforward. As it turns out, however,  $\eta$  is varying considerably with range in a manner dependent on receiver field of view, range to cloud, wavelength, and angular scattering properties. Monte Carlo simulations performed by Platt [51] show that  $\eta$  increases very rapidly with penetration depth from a low value at the base of the cloud to a more or less constant value at optical depths greater than 2–3. The total increase is typically on the order of a factor of 2. In some instances of cirrus clouds, negative values of  $\eta$ , as low as  $-0.6$ , were found at cloud base. This illustrates once more that the interpretation of sole extinction reduction implied by the  $\eta$ -formulation is not correct in general since the backscattering coefficient is also affected. In summary, the correction

algorithms suggested by  $\eta$  can be misleading;  $\eta$  is not a simple factor but a function that varies by a significant amount not only from one application to another but within a same application. Nevertheless, for the specific application of spaceborne cirrus measurements, Wiegner et al. [81] showed that a constant  $\eta$  can provide meaningful correction.

Wandinger [68] made a thorough study of the influence of multiple scattering on Raman and high-spectral-resolution lidar retrieval algorithms. Simulations carried out with the phenomenological model of Weinman [66] showed that errors on the retrieved particle extinction coefficient can be as large as 50% at the base of a cloud, even for fields of view as low as 0.4 mrad (full angle). Generally, the errors drop below 20% with further penetration depth. Backscattering coefficients are less affected with errors much below 20%. Wandinger went on to demonstrate that meaningful corrections of real measurements can be implemented by running direct-problem calculations of multiple scattering contributions. To do this, one needs the size distribution, or the angular scattering function, and the true extinction profile. The size distribution must be assumed, or at least bracketed within realistic limits, because it cannot be inferred from the conventional retrieval tools of the Raman and high-spectral-resolution methods. However, a corrected extinction profile, constrained by the assumed size distribution, can be derived through iterations. The starting solution is the uncorrected extinction  $\alpha_e$  which is inputted along with the assumed size distribution in the chosen multiple scattering model to calculate the corresponding function  $\eta$ . A corrected  $\alpha = \alpha_e / \eta$  is thus obtained and the direct-problem computations are rerun to calculate a new  $\eta$ . The reconstructed  $\alpha_e$  from the last solutions for  $\alpha$  and  $\eta$  is then compared with the measured  $\alpha_e$ . If the differences are greater than preset limits, the  $\alpha$  profile is varied, according to a suitable search algorithm, at the input of the direct-problem model until proper agreement is reached between the reconstructed and measured  $\alpha_e$ 's.

As a follow-on, Reichardt et al. [82] carried out several computations based on the same model of Weinman in search of scaling laws that would allow estimating  $\eta$  from a standardized cloud. Their results show that this is possible, approximately, for such parameters as the transmitter wavelength and the receiver field of view in conditions of near homogeneous clouds. However, simple scaling laws do not seem to work in general.

In summary, corrections or adaptations of single scattering retrieval algorithms to take into account multiple scattering effects are not straightforward. As we have seen, there is a fair number of valid calculation

models of multiple scattering but the main inputs to drive these models are actually the medium properties we wish to correct for. Iterations to derive true values from “effective” values are possible but there is almost always a missing input not available from the retrieval algorithms under study, e.g., the phase function. However, because of the wide range of planned lidar-in-space applications in which multiple scattering cannot be neglected, work is continuing at a steady pace to devise practical retrieval algorithms to account for or even exploit multiple scattering, e.g., Winker [83].

### 3.5 Inverse Problem

Multiple scattering has mostly been considered in the past as a hindrance in single scattering retrieval methods. However, the modeling results of Section 3.3 show that the multiple scattering contributions contain information on particle size that is not available in the single scattering expressions. We review in this section some of the recent efforts made to solve the inverse problem, i.e., to retrieve at least part of this additional information. There are a few promising results but a good deal of theoretical development remains to be done.

#### 3.5.1 Particle Size Distribution

One main characteristic of multiply scattered lidar signals is their dependence on the receiver field of view. This arises because the range of scattering angles contributing to the collected signal widens with the field of view. It is well known that the angular shape of the scattering phase function depends on particle size. At small angles, the scattering is mainly caused by Fraunhofer diffraction which gives a one-to-one relationship with the size of the scatterer. As discussed in Section 3.3, multiple scattering in the conventional lidar geometry of narrow source divergence and receiver field of view is characterized by multiple small-angle forward scatterings. Hence, angularly resolved lidar returns are highly dependent on the phase function forward peak and can in principle be exploited for particle size retrieval.

We outline here one generic approach to solve for the particle size distribution. To keep the inverse problem linear, we assume double scattering only. Hence, the solution we seek will be applicable only to small optical depths which, for clouds of reasonable density, also means

small geometrical depths. We thus make the further assumption that the normalized diameter density distribution,

$$f(\rho) = \frac{1}{N_0(z)} \frac{d}{d\rho} N(z, \rho), \quad (3.114)$$

is independent of the range  $z$ , where  $N(z, \rho)$  is the number of particles of diameter  $\rho$  per unit volume and  $N_0(z)$  is the total number density for all sizes.

Let the returned signal, denoted  $P(z, \Theta_j)$ , be measured at  $\mathcal{M}$  different fields of view, i.e., for  $j = 1, \mathcal{M}$ . We model  $P(z, \Theta_j)$  in the framework of the effective medium theorem of Subsection 3.3.3. Therefore, we have for second-order scatter

$$\begin{aligned} P(z, \Theta_j) = & \frac{K}{z^2} \exp \left[ -2 \int_{z_b}^z \alpha(z') dz' \right] \int_{z_b}^z dz' 2\alpha_s(z') \\ & \times \int_0^{\phi_j(z')} 2\pi \sin \phi p(\phi) \alpha_{sb} p_b(z, \pi - z'\phi/z) d\phi, \end{aligned} \quad (3.115)$$

where  $K$  is the instrument constant,  $z_b$  is the range to cloud base,  $p(\phi)$  is the phase function for forward scattering assumed independent of  $z$ ,  $\alpha_{sb} p_b(z, \pi - z'\phi/z)$  is the angular backscattering function responsible for the single backscattering event, and

$$\tan \phi = \frac{z}{z - z'} \tan \Theta. \quad (3.116)$$

We take  $\alpha_{sb} p_b$  different from  $\alpha_s p$  to include Raman lidar, high-spectral-resolution lidar, reflection from ground or sea surface, etc.

Because our objective is to solve for the particle size distribution, we express  $p(\phi)$  as a sum over the individual diameters of the distribution, i.e.,

$$p(\phi) = \frac{1}{4} \int_{\rho_{min}}^{\rho_{max}} d\rho f(\rho) \frac{\pi \rho^2}{\langle \sigma_s \rangle} Q_s(\lambda, \rho, m) \mathcal{P}(\phi, \lambda, \rho, m), \quad (3.117)$$

where  $Q_s(\lambda, \rho, m)$  and  $\mathcal{P}(\phi, \lambda, \rho, m)$  are, respectively, the scattering efficiency and phase function at the wavelength  $\lambda$  for a particle of diameter  $\rho$  and refractive index  $m$ , and  $\langle \sigma_s \rangle$  is the average particle scattering cross-section given by

$$\langle \sigma_s \rangle = \frac{1}{4} \int_{\rho_{min}}^{\rho_{max}} d\rho f(\rho) \pi \rho^2 Q_s(\lambda, \rho, m). \quad (3.118)$$



We do not write  $p_b(z, \pi - z'\phi/z)$  in terms of  $f(\rho)$  because it does not necessarily arise from the same particle distribution as indicated above. Actually, we consider  $p_b(z, \pi - z'\phi/z)$  as a known function. In cases where  $p_b = p$ , we assume as a first approximation that  $p_b \simeq \text{constant}$  near  $180^\circ$ . For aerosol particles large compared with the wavelength for which this method is applicable, the magnitude of the phase function variations is much less in the near backward direction than in the near forward direction. The smaller backward variations can subsequently be taken into account by iterations in which  $p_b(z, \pi - z'\phi/z)$  is evaluated from the  $f(\rho)$  determined on the preceding iteration cycle.

We approximate the integral over  $\rho$  resulting from Eq. (3.117) as a discrete sum of  $\mathcal{M}$  terms as follows:

$$\int_{\rho_{\min}}^{\rho_{\max}} d\rho f(\rho) \cdots = \sum_{i=1}^{\mathcal{M}} \int_{\rho_i}^{\rho_{i+1}} d\rho f(\rho) \cdots = \sum_{i=1}^{\mathcal{M}} \bar{f}_i \int_{\rho_i}^{\rho_{i+1}} d\rho \cdots, \quad (3.119)$$

where  $\bar{f}_i$  is the average  $f$  over the  $i$ th interval. Within the intervals, we keep the integration over  $\rho$  because the functions  $Q_s$  and  $\mathcal{P}$  can oscillate rapidly with  $\rho$ . Using Eqs. (3.117)–(3.119), we rewrite (3.115) as a linear equation

$$P_j = \bar{f}_i A_{ij}, \quad (3.120)$$

where

$$P_j = P(z, \Theta_{j+1}) - P(z, \Theta_j), \quad (3.121)$$

$$A_{ij} = C(z) \int_{\rho_i}^{\rho_{i+1}} d\rho \rho^2 \int_{z_b}^z dz' \alpha_s(z') \int_{\phi_j(z')}^{\phi_{j+1}(z')} \sin \phi \times Q_s(\lambda, \rho, m) \mathcal{P}(\phi, \lambda, \rho, m) p_b(z, \pi - z'\phi/z) d\phi, \quad (3.122)$$

$$C(z) = \frac{\pi^2}{\langle \sigma_s \rangle} \frac{K}{z^2} \alpha_{sb}(z) \exp \left[ -2 \int_{z_b}^z \alpha(z') dz' \right]. \quad (3.123)$$

Equation (3.120) is a linear equation that can be inverted for  $\bar{f}_i$  with well-known constrained or regularized techniques [84] once the matrix elements of  $A_{ij}$  are calculated. The  $\alpha_s(z')$  that enters the definition of  $A_{ij}$  can be estimated from an assumed value of the single scattering albedo and a backward Klett's [6] solution which becomes independent of the far end boundary value at the small penetration depths for which the

second-order expression (3.115) was derived. The elements of  $A_{ij}$  are only known to a multiplicative constant because of the unknown  $\langle\sigma_s\rangle$ ,  $K$  and  $\alpha_{sb}$ . Therefore,  $\bar{f}_i$  can be solved only in relative units. However, because of the Klett's solution for  $\alpha$ , the particle number density can be estimated a posteriori.

The matrix elements  $A_{ij}$  can be obtained either with the exact Mie-calculated  $Q_s(\lambda, \rho_i, m)$  and  $\mathcal{P}(\phi, \lambda, \rho_i, m)$  in the case of spherical particles, or with the Fraunhofer expression for size parameters  $2\pi\rho_i/\lambda \gg 1$  since the significant contributions come from small-angle scatterings. Because of its simple analytic form, the Fraunhofer formula is useful in either case to select the  $\rho$  and  $\Theta$  bin sizes that make the diagonal elements  $A_{ii}$  of comparable magnitude.

Illustrative measurements and inversion results obtained by Roy et al. [34] in laboratory-controlled water droplet clouds are plotted in Fig. 3.8. The lidar returns were measured in the polarization direction perpendicular to the source polarization. This had the advantage of making  $p_b(z, \pi) \simeq 0$  and, thus, eliminating the large single-scattering contribution. Results obtained at two optical depths,  $\gamma = 0.2$  and  $0.4$ , respectively, are shown. The agreement is good in the case of the smaller

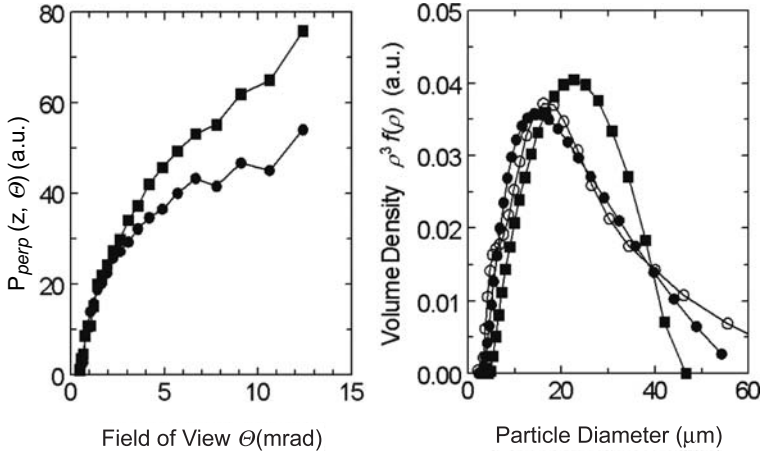


**Fig. 3.8.** Lidar MFOV measurements (left) and corresponding retrieved droplet volume density distributions  $\rho^3 f(\rho)$  compared with *in situ* particle-size data (right).  $P_{\text{perp}}$  is the collected return in the polarization direction perpendicular to that of the linearly polarized laser source. The scattering medium is a water droplet cloud produced in a 22-m long  $\times$  2.4-m  $\times$  2.4-m wide chamber located 100 m from the lidar. Solid circles: penetration depth of 6 m and  $\gamma = 0.2$ ; solid squares: penetration depth of 10 m and  $\gamma = 0.4$ ; and open circles: *in situ* measurements.

optical depth but only fair at  $\gamma = 0.4$ . This is consistent with the validity domain of the second-order approximation made in the derivation of the linear equation (3.120). In both cases, the lidar-retrieved distribution cuts off at smaller diameters than the *in situ* measurements. The probable cause is the limited field-of-view range of the instrument for the given measurement geometry. For example, large droplets at far distances require very narrow fields of view whereas small particles at short distances require wide fields of view as it can be easily inferred from Eq. (3.116). For the results of Fig. 3.8, the smallest fields of view are still too wide for resolving the narrow angular spreading due the large droplets, hence the rapid cutoff of the retrieved distributions at the large-diameter end. Following the same geometrical-factor argument, Roy et al. [85] have suggested that a spaceborne MFOV lidar receiver of reasonable angular aperture could be used, because of the long ranges, to characterize the size of submicrometer aerosol particles in the atmospheric boundary layer.

An earlier application of a second-order scattering size retrieval method was published by Benayahu et al. [86]. They used two receivers in a bistatic configuration. One receiver had its axis shifted from the beam axis so as not to collect singly scattered radiation. The idea is interesting but the chosen instrument parameters and the measurement geometry made the smallest scattering angle  $\phi$  contributing to the multiple scattering channel of the order of  $10^\circ$ . This is too large to collect enough of the diffraction scatterings from typical cloud droplets. As a result, the sensitivity on droplet size is weak. A sample set of inversion results obtained for marine stratus clouds yielded an effective droplet diameter  $\langle \rho^3 \rangle / \langle \rho^2 \rangle \simeq 6.6 \mu\text{m}$ , which is 3–4 times smaller than most published measurement values for marine clouds.

Figure 3.8 shows that size information is indeed retrievable from experimental multiply scattered lidar data but the existing retrieval methods are still limited in their application range. The results obtained to date are from early initiatives and better performances are expected with future developments. There is currently ongoing research to derive more robust methods for application to cirrus clouds, e.g., Eloranta [87].

### 3.5.2 Extinction and Effective Particle Size

It is now well established that the multiply scattered lidar returns are functions of the medium extinction coefficient and angular scattering properties or particle size distribution. However, there are no simple

mathematical relations between these local properties and the observables for multiple scattering of order greater than 2. Actually, the observables are the results of many integrated interactions as illustrated by the multiple integrals of the phenomenological models of Subsection 3.3.2. This makes the inverse problem of retrieving the local extinction coefficient and particle size particularly difficult.

It has been proposed to use the powerful direct-problem tools to carry out multi-dimensional searches of the medium parameters that would reproduce the measured signals. For example, Oppel et al. [88] have performed retrieval simulations for a medium of uniform extinction and size distribution. The assumed measured signals were the parallel and perpendicularly polarized returns. The chosen calculation algorithm was a variance-reduction Monte Carlo model. The search grid for that example was two-dimensional and made up of the extinction coefficient and the modal radius of the size distribution; the distribution was assumed to be of the Deirmendjian's C1 type. The convergence criterion was the minimization of the root-mean-square distance between the calculated and the "measured" signals. Oppel et al. [88] showed that an adaptive step size random search was significantly more efficient than a uniform search. They also found that the sensitivity of the minimization search is good for the extinction coefficient but low for the modal radius. The search-based inversion is conceptually simple and adaptable to different kinds of retrieval problems. However, it requires extensive computational resources and its practicality becomes questionable for non-uniform media.

The concept of using direct-problem calculation methods in the inverse problem is also applicable to iterative solution algorithms. To fix ideas, let the multiple scattering lidar equation be written

$$P(z, \Theta) = P_{ss}(z)M(z, \Theta) = P_{ss}(z)[1 + F_d(z, \Theta) + F_g(z, \Theta)], \quad (3.124)$$

where  $\Theta$  is the half-angle receiver field of view,  $P_{ss}$  is the conventional single scattering lidar expression given by Eq. (3.1), and  $M(z, \Theta)$  is the multiple-to-single scattering ratio split into components  $F_d$  and  $F_g$ , respectively, for diffraction scatterings alone and for all other scatterings that involve at least one geometrical optics scattering. Geometrical optics means refraction and reflection. The QSA approximation of multiple small-angle forward scatterings and a single large-angle backscattering is implied in Eq. (3.124). The separation of  $M$  into  $F_d$  and  $F_g$  has the advantage of consolidating the dependence on particle size almost exclusively in  $F_d$ .

The iteration principle is rather simple. If an intermediate solution for the extinction coefficient and the particle size is known, the multiple-to-single scattering ratio  $M$  can be calculated by direct-problem methods to correct the measured functions used in the single scattering inversion algorithms, thus allowing the calculation of a refined solution. For example, the Klett algorithm, Eq. (3.7), could be rerun after redefining the signal function  $S(z)$  as follows:

$$S(z) = \frac{P(z, \Theta)}{M(z, \Theta)} \frac{z^2}{k(z)}, \quad (3.125)$$

where  $P(z, \Theta)$  is the measured multiply scattered lidar return, and  $M(z, \Theta)$  and  $k(z) = \beta(z)/\alpha(z)$  are the multiple-to-single scattering and the backscatter-to-extinction ratios calculated with the intermediate solutions for extinction and particle size. It is assumed in Eq. (3.125) that the instrument function  $K$  is independent of  $z$ . Needed to implement such an iteration method are, in addition to a direct-problem calculation model for  $M(z, \Theta)$ , an algorithm for particle size and an initialization procedure.

Veretennikov et al. [89] use the iteration concept described above to retrieve cloud extinction profiles from actual multiple-field-of-view (MFOV) lidar measurements. They avoid size retrieval by fixing the particle effective radius. They initialize their extinction solution either on a reference value obtained with the slope method in cases where an interval of constant extinction can be identified, or by the use of a regularization constant in lieu of a boundary value in the denominator of the Klett expression (3.7). They compare retrievals obtained for different fields of view and show that the iteration is generally stable. The retrieved extinction profiles are in good agreement with what is expected from continental stratus clouds but no independent measurements were available.

There is no rigorous, general method of determining simultaneously the extinction coefficient and the effective or average particle diameter in optically dense media. The difficulty resides with the size retrieval. The particle size dependence of multiply scattered lidar returns is a complex integrated effect and we are missing a robust analytic inverse relation between the measured returns and the local particle size distribution. One preliminary semiempirical iterative technique has been described by Bissonnette et al. [90].

The size retrieval algorithm proposed by Bissonnette et al. consists in measuring the field-of-view spread of the multiply scattered returns,

quantifying this spread by a characteristic scale, and relating this scale to the width of the forward peak of the scattering phase function or the effective particle diameter, by means of a semiempirical model. The existence of a relation between the field-of-view scale and the particle size rests on the now well-established premise that, for a receiver footprint less than the scattering mean free path and for optical depth less than 3–4, the field-of-view dependence of  $P(z, \Theta)$  is mostly driven by the small-angle forward scatterings. The implementation of the method requires a direct-problem calculation model of the multiple scattering function  $M(z, \Theta)$  in Eq. (3.125) to run the iterations and an initialization procedure.

The proposed initialization procedure is based on the approximations that at the onset of multiple scattering the receiver largest field of view  $\Theta_{max}$  is sufficiently wide to encompass all diffraction forward scatterings and yet narrow enough for  $P(z, \Theta)$  not to be significantly affected by the geometrical optics scatterings, and that the smallest field of view  $\Theta_{min}$  is small enough for  $P(z, \Theta_{min})$  to approach the single-scattering return  $P_{ss}(z)$ . Under these conditions, it is easy to show, for instance using Eloranta's analytical model [61], that

$$\gamma(z) \simeq \ln \left[ 1 + \frac{1}{\delta_b} \frac{P(z, \Theta_{max}) - P(z, \Theta_{min})}{P(z, \Theta_{min})} \right], \quad (3.126)$$

where  $\delta_b$  is the ratio of the backscatter coefficient averaged over  $\Theta_{max}$  to the single-scattering coefficient.  $\delta_b$  arises because the backscattering function  $p_b$  is not necessarily uniform near  $180^\circ$ . For Raman and high-spectral-resolution lidars,  $\delta_b$  is conveniently equal to unity. For elastic backscattering in low-level water clouds probed from the ground, we have  $\delta_b \simeq 0.7$ . Monte Carlo simulations show that the approximation (3.126) is satisfied to within 20% in the interval

$$1.2 \leq P(z, \Theta_{max})/P(z, \Theta_{min}) \leq 1.5. \quad (3.127)$$

Therefore, the optical depth in the range defined by (3.127) can be determined from the relative strengths of the returns measured at  $\Theta_{min}$  and  $\Theta_{max}$ , independently of a calibration constant. This  $\gamma(z)$  was successfully used in Ref. [90] to initialize the iterations for the simultaneous retrieval of the extinction coefficient and the effective particle diameter. A Klett solution applied to the modified signal  $S(z)$  of Eq. (3.125) was used for the extinction coefficient and a semiempirical model for the particle diameter. The direct-problem model needed to calculate  $M(z, \theta)$  can

be any model that accepts the extinction and particle diameter profiles obtained from the previous-iteration solutions as inputs.

Tests on Monte-Carlo-simulated returns reported in Ref. [90] show that the retrieved extinction coefficients are within 3–4% of the true values. The performance is not as good for the particle diameter. While the solutions agree very well with the true effective diameters on average, the fluctuations are of the order of  $\pm 25\%$ . In addition, there are limits on the retrievable sizes. This does not result from flaws in the solution method but rather from the limited angular resolution of the MFOV receiver: the field-of-view scale cannot be determined if it falls outside of the interval  $[\theta_{min}, \theta_{max}]$ .

Comparisons of lidar retrievals with actual field data are also shown in Ref. [90]. The experiment consisted of vertical lidar probings from a fixed ground site and simultaneous *in situ* aircraft measurements in low stratus clouds. Because of the variable and large separation (5–40 km) between the lidar and the aircraft, the comparisons were performed on long time averages only. The results show good correlation between the lidar solutions and the aircraft measurements of the cloud liquid water content (LWC) and effective droplet diameter. However, there is a bias—lidar underestimation—varying between 15% and 30% depending on the aircraft sensor. The analysis shows that the discrepancy cannot be attributed to the lidar alone.

Although there is still much analytical development to be made, the results of Ref. [90] demonstrate that multiple-scattering-based lidar retrieval can work. One advantage of multiple scattering retrieval is the information gained on particle size which allows extrapolation to secondary products like cloud liquid water content (LWC), extinction at other wavelengths, cloud radiative properties, etc. The applications are limited to measurement geometries and medium properties that satisfy the QSA approximation. Optical depths are limited to 3–4, which fortunately corresponds to practical hardware limits, and the bounds on particle size are determined by the measurement geometry and the transceiver technical specifications. Developments in gateable intensified imaging receivers could broaden the scope of MFOV detection.

### 3.5.3 Bulk Properties

The diffusion limit of multiple scattering offers real opportunities of extending the usefulness of lidars. We have seen that the QSA

approximation, which constitutes the basis of the inversion methods described so far in this section, is no longer valid when the lidar footprint exceeds the scattering mean free path. A practical situation is that of a spaceborne lidar probing water clouds in the atmospheric boundary layer. However, there are still measurable reflected photons in such instances and they return information on cloud properties. For dense clouds, the diffusion results of Eqs. (3.110)–(3.113) show that the cloud physical and optical thicknesses and their average scattering properties, defined by the asymmetry factor, are in principle retrievable from space-time measurements of the reflected aureole.

Measuring the off-beam aureole poses some technical challenges. Unusually wide fields of view are required which complicates the rejection of background radiation in favor of the faint diffusion-reflected signals. Two experimental systems have been built and tested so far: a high-speed gated/intensified imaging camera with a  $60^\circ$  full-angle field of view for ground-based use [78], and a fiber bundle 8-ring MFOV receiver with a  $6^\circ$  angular aperture for airborne use [91]. In the first case, Love et al. [78] obtained time resolved images from a stratus cloud deck at 1 km above ground, clearly showing a “wave” of diffusing light propagating radially from the central impact point of the laser source pulse; these authors used pairs of modified versions of Eqs. (3.111)–(3.113) to estimate the geometrical and optical thicknesses of the cloud deck. In the second case, Cahalan et al. [91] carried out preliminary measurements with the MFOV airborne lidar that reveal time delays between the outer- and central-ring signals from a thick stratus layer 7 km away; their results are compatible with the diffusion model of Eqs. (3.110)–(3.113). Finally, it is noteworthy that the LITE data in Fig. 3.5 have been used in Eqs. (3.111) and (3.112) to infer with reasonable accuracy the optical depth and physical thickness of the marine stratocumulus deck from which the LITE returns originated [92].

Considering the growing interest [93] in space-based remote sensing of clouds, rapid developments are expected. Diffusion retrieval may become in the future a significant contributor in the remote characterization of thick water clouds.

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## Lidar and Atmospheric Aerosol Particles

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### 4.1 Introduction

Atmospheric aerosols play an important role in many atmospheric processes. Although only a minor constituent of the atmosphere, they have appreciable influence on the Earth's radiation budget, air quality and visibility, clouds, precipitation, and chemical processes in the troposphere and stratosphere. The occurrence, residence time, physical properties, chemical composition, and corresponding complex-refractive-index characteristics of the particles, as well as the resulting climate-relevant optical properties are subject to large diversity especially in the troposphere because of widely different sources and meteorological processes. Therefore, vertically resolved measurements of physical and optical properties of particles such as the particle surface-area concentration, volume and mass concentrations, mean particle size, and the volume extinction coefficient are of great interest. Routine (long-term), height-resolved observations of these parameters can only be carried out with lidar.

Commonly, aerosols are described in terms of aerosol types in climate models. These aerosol types are defined as internal or external mixtures of different components, and each component has distinctive properties. The *water-insoluble* part of aerosol particles consists mostly of soil particles with some amount of organic material. The *water-soluble* part originates from gas-to-particle conversion. It consists of various kinds of sulfates, nitrates, and other water-soluble substances, which also include organics. *Soot* represents absorbing black carbon. *Sea-salt* particles represent the various kinds

of salt contained in seawater. *Mineral* particles describe desert dust produced in arid regions. The *mineral-transported* component describes desert dust that is transported over long distances, which leads to the depletion of large particles. *Sulfate droplets* are used to describe sulfate found in Antarctic aerosol and the stratospheric background aerosol.

Table 4.1 lists some characteristics of important aerosol types used in climate modeling. These aerosol types differ in their relative contribution of the various aerosol components. A detailed overview can be found elsewhere [1, 3]. On the one hand, these types span the average conditions; on the other hand, they take account of extreme conditions for sensitivity studies. It has to be observed that actual measurements may show aerosol properties significantly different from these aerosol types.

The three different types of continental aerosols differ in their amount of soot, which is considered as a parameter describing the influence of anthropogenic activities. Furthermore, the overall concentration of particles strongly increases from continental clean to continental polluted conditions. The urban aerosol describes strong pollution in urban areas. It has the highest concentration of particles, in particular, that

**Table 4.1.** Properties of aerosol types [1]<sup>a</sup>

| Aerosol type               | $N$<br>( $\text{cm}^{-3}$ ) | $r_{\text{eff}}$<br>( $\mu\text{m}$ ) | $ssa$<br>( $0.55\ \mu\text{m}$ ) | $g$<br>( $0.55\ \mu\text{m}$ ) | $\tilde{a}$<br>( $0.35\text{--}0.55\ \mu\text{m}$ ) | $\tilde{a}$<br>( $0.55\text{--}0.8\ \mu\text{m}$ ) |
|----------------------------|-----------------------------|---------------------------------------|----------------------------------|--------------------------------|-----------------------------------------------------|----------------------------------------------------|
| Cont. clean                | 2600                        | 0.247                                 | 0.972                            | 0.709                          | 1.10                                                | 1.42                                               |
| Cont. average              | 15,300                      | 0.204                                 | 0.925                            | 0.703                          | 1.11                                                | 1.42                                               |
| Cont. polluted             | 50,000                      | 0.150                                 | 0.892                            | 0.698                          | 1.13                                                | 1.45                                               |
| Urban                      | 158,000                     | 0.139                                 | 0.817                            | 0.689                          | 1.14                                                | 0.43                                               |
| Desert                     | 2300                        | 1.488                                 | 0.888                            | 0.729                          | 0.20                                                | 0.17                                               |
| Marit. clean               | 1520                        | 0.445                                 | 0.997                            | 0.772                          | 0.12                                                | 0.08                                               |
| Marit. polluted            | 9000                        | 0.252                                 | 0.975                            | 0.756                          | 0.41                                                | 0.35                                               |
| Marit. tropical            | 600                         | 0.479                                 | 0.998                            | 0.774                          | 0.07                                                | 0.04                                               |
| Arctic                     | 6600                        | 0.120                                 | 0.887                            | 0.721                          | 0.85                                                | 0.89                                               |
| Antarctic                  | 43                          | 0.260                                 | 1.000                            | 0.784                          | 0.34                                                | 0.73                                               |
| Stratosphere<br>(12–35 km) | 3                           | 0.243                                 | 1.000                            | 0.784                          | 0.74                                                | 1.14                                               |

<sup>a</sup>Number concentration is denoted by  $N$ . The effective radius  $r_{\text{eff}}$  describes the mean size of the particle ensemble. The single-scattering albedo  $ssa$  is defined as the ratio of total scattering to extinction of the investigated particle ensemble. The asymmetry parameter  $g$  is a measure of light scattered toward the forward direction compared with the light scattered toward the back direction. The Ångström exponent  $\tilde{a}$  [2] describes the spectral slope of the optical coefficients. All numbers hold for a relative humidity of 80%. Effective radius is calculated for 50% relative humidity. A further discussion of some of the parameters is given in Section 4.3.

of soot. Desert aerosol describes the conditions over desert regions without distinguishing between local properties. There are three types of marine aerosols, which contain a different concentration of sea salt depending on the actual wind speed, and soot, which reflects the anthropogenic influence on the maritime environment. Arctic aerosol consists of particles transported from the mid-latitude continental areas and therefore contains soot. The Antarctic aerosol type exhibits a mixture of mostly sulfate droplets with some amount of sea salt and mineral particles. The stratospheric aerosol is given for background conditions.

Elastic-backscatter lidars have extensively been used to investigate clouds and aerosol layers since the early 1960s when Fiocco and Grams [4] reported the first lidar-derived stratospheric aerosol profiles. Only in recent years, however, has significant progress been made toward a quantitative study of atmospheric aerosol properties solely based on lidar. Aerosol lidars were involved in large, integrated aerosol field campaigns such as the Tropospheric Aerosol Radiative Forcing Observational Experiment (J. Geophys. Res. **104**, D2, 1999 and **105**, D8, 2000), the Aerosol Characterization Experiment 2 (Tellus **52B**, No. 2, 2000), the Indian Ocean Experiment (J. Geophys. Res. **106**, D22, 2001 and **107**, D19, 2002), the Lindenberg Aerosol Characterization Experiment (J. Geophys. Res. **107**, D21, 2002), and the Asian Pacific Regional Aerosol Characterization Experiment (J. Geophys. Res. **108**, D23, 2003 and **109**, D19, 2004). These so-called aerosol closure experiments were conducted to study the impact of anthropogenic particles on the climate system. Networks of aerosol lidars such as the Asian Dust Network [5] and the European Aerosol Research Lidar Network [6] were established to investigate the horizontal and vertical distribution of natural and anthropogenic aerosol plumes in a coherent way on a regional to continental scale. The US National Aeronautics & Space Administration (NASA) and, in cooperation, the European Space Agency (ESA) and the National Space Development Agency of Japan (NASDA) will launch satellite-borne lidars for a multiyear mapping of global aerosol distributions and for the characterization of the long-range transport of particles.

In this chapter we review and critically discuss the two most important methods for the determination of optical particle parameters from lidar observations and the techniques that are used to retrieve physical properties of tropospheric and stratospheric aerosols. In Subsection 4.2.1 the technique is explained that is taken to compute the particle backscatter coefficient (scattering coefficient at  $180^\circ$ , normalized to the unit solid

angle) from return signals measured with the widely used elastic or standard backscatter lidar [7–10]. Elastic-backscatter lidars detect the total atmospheric backscatter without separation of particle and molecular contributions to the backscattered signal. The main drawback of this method is that trustworthy profiles of the climate-relevant volume extinction coefficient of the particles cannot be obtained. The extinction profile must be estimated from the determined backscatter-coefficient profile.

By applying the so-called Raman-lidar technique [11, 12] or the high spectral resolution lidar (HSRL) method [13–15] the profile of the particle extinction coefficient can directly be determined. In addition, the profile of the backscatter coefficient is obtained. An aerosol Raman lidar or an HSRL measures two signal profiles, which permit the separation of particle and molecular backscatter contributions. This method is described in Subsection 4.2.2.

For completeness, the scanning or multiangle lidar technique [16–19] is another technique to derive vertical profiles of the particle extinction coefficient. The most critical requirement here is the need for horizontally homogeneous particle backscattering and extinction at all measurement heights. This condition is often not fulfilled, at least not in the convective boundary layer.

Simultaneously measured extinction and backscatter coefficient profiles at several wavelengths between 300 and 1100 nm are the fundamental prerequisite for a successful, accurate retrieval of physical properties of tropospheric particles from the optical ones [20]. Tropospheric aerosols over the continents often contain a complex mixture of natural (marine and dust particles) and anthropogenic particles (mainly sulfate and soot particles) so that the refractive-index characteristics are unknown. Furthermore, because of the great variability of sources and because of coagulation, mixing, transport, and removal processes, the size distribution that covers the particle diameter range from a few nanometers to several micrometers often shows a complex, multimodal shape. The basic methodology of the inversion technique applied to tropospheric lidar observations is explained in Subsection 4.3.1.

Stratospheric aerosol conditions are comparatively simple. As a consequence, much simpler retrieval schemes can be applied here to determine the microphysical properties from lidar data. Sulfuric-acid/water droplets form the stratospheric aerosol layer. For these particles the refractive index is accurately known. The size distribution can well be described by monomodal logarithmic-normal distributions

under background conditions. A second mode is present during several years after major volcanic eruptions. Temporal changes in the size characteristics can be considered to be very slow compared with tropospheric variations. Lidar methods for the retrieval of microphysical properties of stratospheric particles are discussed in Subsection 4.3.2. A few measurement examples illustrating the potential of modern aerosol lidars are presented in Section 4.4.

## 4.2 Determination of Optical Parameters of Atmospheric Particles

### 4.2.1 Elastic-Backscatter Lidar

The lidar equation for return signals due to elastical backscatter by air molecules and aerosol particles can, in its simplest form, be written as [7]

$$P(R) = \frac{E_0 \eta_L}{R^2} O(R) \beta(R) \exp \left[ -2 \int_0^R \alpha(r) dr \right]. \quad (4.1)$$

$P(R)$  is the signal owing to Rayleigh and particle scattering received from distance  $R$ ,  $E_0$  is the transmitted laser pulse energy,  $\eta_L$  contains lidar parameters describing the efficiencies of the optical and detection units, and  $O(R)$  describes the overlap between the outgoing laser beam and the receiver field of view.  $\beta(R)$  (in  $\text{km}^{-1} \text{sr}^{-1}$ ) and  $\alpha(R)$  (in  $\text{km}^{-1}$ ) are the coefficients for backscattering and extinction, respectively. Backscattering and extinction are both caused by particles (index aer) and molecules (index mol):

$$\beta(R) = \beta_{\text{aer}}(R) + \beta_{\text{mol}}(R), \quad (4.2)$$

$$\alpha(R) = \alpha_{\text{aer}}(R) + \alpha_{\text{mol}}(R). \quad (4.3)$$

Molecular absorption effects are ignored. These effects have to be removed from the measured signals before applying the methods presented in this chapter.

Equations (4.1)–(4.3) can be summarized to

$$S(R) = E_0 \eta_L [\beta_{\text{aer}}(R) + \beta_{\text{mol}}(R)] \exp \left[ -2 \int_0^R [\alpha_{\text{aer}}(r) + \alpha_{\text{mol}}(r)] dr \right] \quad (4.4)$$



with the range-corrected lidar signal  $S(R) = R^2 P(R)$ . The overlap is assumed to be complete [ $O(R) \equiv 1$ ], i.e., the minimum distance  $R_{\min}$  at which measurements can be made may be defined by  $O(R) \leq 1$  for  $R \leq R_{\min}$ . The molecular scattering properties,  $\beta_{\text{mol}}(R)$  and  $\alpha_{\text{mol}}(R)$ , can be determined from the best available meteorological data of temperature and pressure or approximated from appropriate standard atmospheres so that only the aerosol scattering and absorption properties,  $\beta_{\text{aer}}(R)$  and  $\alpha_{\text{aer}}(R)$ , remain to be determined.

In the next step we introduce the particle extinction-to-backscatter ratio (lidar ratio)

$$L_{\text{aer}}(R) = \frac{\alpha_{\text{aer}}(R)}{\beta_{\text{aer}}(R)} \quad (4.5)$$

in analogy to the molecular lidar ratio

$$L_{\text{mol}} = \frac{\alpha_{\text{mol}}(R)}{\beta_{\text{mol}}(R)} = \frac{8\pi}{3} \text{ sr}. \quad (4.6)$$

In contrast to the molecular lidar ratio, the particle lidar ratio is range-dependent because it depends on the size distribution, shape, and chemical composition of the particles.

In addition, we introduce the term

$$Y(R) = L_{\text{aer}}(R)[\beta_{\text{aer}}(R) + \beta_{\text{mol}}(R)]. \quad (4.7)$$

In Sasano et al. [10]  $Y(R)$  is expressed as a function of  $\alpha_{\text{aer}}(R)$ .

However, we prefer Eq. (4.7) because the primary information in the measured elastic lidar returns [cf. Eq. (4.1)] is the backscatter coefficient under typical tropospheric conditions with particle vertical optical depth of  $\leq 0.3$  in the visible spectrum around 550 nm. Under these conditions only the backscatter coefficient can be derived with good accuracy from the elastic backscatter signal.

After substituting  $\alpha_{\text{aer}}(R)$  and  $\alpha_{\text{mol}}(R)$  in Eq. (4.4) with the expressions (4.5) and (4.6) and inserting  $Y(R)$  from Eq. (4.7), the resulting equation can be written as

$$\begin{aligned} S(R)L_{\text{aer}}(R) \exp \left\{ -2 \int_0^R [L_{\text{aer}}(r) - L_{\text{mol}}] \beta_{\text{mol}}(r) dr \right\} \\ = E_0 \eta_L Y(R) \exp \left[ -2 \int_0^R Y(r) dr \right]. \end{aligned} \quad (4.8)$$

Taking the logarithms of both sides of Eq. (4.8) and differentiating them with respect to  $R$  gives

$$\begin{aligned} & \frac{d \ln \left( S(R) L_{\text{aer}}(R) \exp \left\{ -2 \int_0^R [L_{\text{aer}}(r) - L_{\text{mol}}] \beta_{\text{mol}}(r) dr \right\} \right)}{dR} \\ &= \frac{1}{Y(R)} \frac{dY(R)}{dR} - 2Y(R). \end{aligned} \quad (4.9)$$

Finally, we solve Eq. (4.9), known as the Bernoulli equation, for the boundary condition

$$Y(R_0) = L_{\text{aer}}(R_0) [\beta_{\text{aer}}(R_0) + \beta_{\text{mol}}(R_0)] \quad (4.10)$$

to obtain [10]:

$$\begin{aligned} & \beta_{\text{aer}}(R) + \beta_{\text{mol}}(R) \\ &= \frac{S(R) \exp \left\{ -2 \int_{R_0}^R [L_{\text{aer}}(r) - L_{\text{mol}}] \beta_{\text{mol}}(r) dr \right\}}{\frac{S(R_0)}{\beta_{\text{aer}}(R_0) + \beta_{\text{mol}}(R_0)} - 2 \int_{R_0}^R L_{\text{aer}}(r) S(r) T(r, R_0) dr}, \quad \text{with} \\ & T(r, R_0) = \exp \left\{ -2 \int_{R_0}^r [L_{\text{aer}}(r') - L_{\text{mol}}] \beta_{\text{mol}}(r') dr' \right\}. \end{aligned} \quad (4.11)$$

The profile of the particle extinction coefficient can be estimated from the solution  $\beta_{\text{aer}}(R)$  by

$$\alpha_{\text{aer}}(R) = L_{\text{aer}}(R) \beta_{\text{aer}}(R). \quad (4.12)$$

Equation (4.11) can, in principle, be integrated by starting from the reference range  $R_0$ , which may be either the near end ( $R > R_0$ , forward integration) or the remote end ( $R < R_0$ , backward integration) of the measuring range. Numerical stability, which is not to be mistaken for accuracy, is, however, given only in the backward integration case [8]. This fundamental formalism used to analyze elastic-backscatter lidar data originates from Hitschfeld and Bordan's [21] radar application. However, the technique is often referred to as the Klett method, as Klett [8] introduced the backward integration scheme and restated in this way the Bernoulli solution in a very convenient form for the analysis of lidar observations.

The reference range  $R_0$  in Eq. (4.11) is usually chosen such that at  $R_0$  the particle backscatter coefficient is negligible compared to the

known molecular backscatter value. Such clear-air conditions are normally given in the upper troposphere for laser wavelengths  $\leq 700$  nm. Note that Rayleigh scattering is proportional to  $\lambda^{-4}$  and thus strongly depends on the wavelength of the transmitted laser pulse.

The most critical input parameter in the Klett method is the particle lidar ratio  $L_{\text{aer}}(R)$ . As mentioned, this quantity depends on the microphysical, chemical, and morphological properties of the particles. All of these properties, in turn, depend on relative humidity. The lidar ratio can vary strongly with height, especially when marine, anthropogenic (urban, biomass burning), and/or desert dust particles or mixtures of these basic aerosol types are present in layers above each other [22, 23]. Typical lidar ratios of the mentioned different aerosol types as measured with our Raman lidar at 532 nm are given in Table 4.2. Variations between about 20 and 100 sr make it practically impossible to estimate trustworthy extinction profiles from Eq. (4.12). Even in the well-mixed layer, the lidar ratio is not constant with height because relative humidity increases with height. In cases with accompanying sun photometer observations that deliver the optical depth (integral over the extinction coefficient profile), a column-related lidar ratio can be estimated from the ratio of the optical depth to the column-integrated backscatter coefficient determined from Eq. (4.11). This lidar ratio can only be considered as a first guess, the true lidar ratio profile remains unknown.

A long-lasting discussion of achievements and limitations of the lidar method applied may be found in the literature [7–10, 12, 24–29]. The procedure, with all its subsequent modifications and improvements, simply suffers from the fact that two physical quantities, the particle backscatter coefficient and the particle extinction coefficient, must be determined from only one measured quantity, the elastic lidar return.

#### 4.2.2 Raman Lidar and HSRL

This unsatisfactory situation improved significantly when the first Raman lidar experiments demonstrated that accurate vertical profiling

**Table 4.2.** Typical lidar ratios for different aerosol types at 532 nm wavelength determined with a Raman lidar

|                                                   |           |
|---------------------------------------------------|-----------|
| Marine particles [22, 30]                         | 20–35 sr  |
| Saharan dust [31]                                 | 50–80 sr  |
| Less absorbing urban particles [22, 30]           | 35–70 sr  |
| Absorbing particles from biomass burning [30, 32] | 70–100 sr |

of the particle extinction coefficient throughout the entire troposphere is possible [11, 12]. After the Pinatubo eruption in 1991 it was immediately shown that even at stratospheric heights profiles of the volume scattering coefficient can easily be obtained with ground-based Raman lidars [33–36]. First attempts to infer particle extinction properties from Raman signal profiles were reported by Gerry and Leonard [37]. First accurate horizontal transmission measurements with Raman lidar were done by Leonard and Caputo [38].

Two types of lidars for extinction profiling are available. The mentioned Raman lidar measures lidar return signals elastically backscattered by air molecules and particles and inelastically (Raman) backscattered by nitrogen and/or oxygen molecules (cf. Chapter 9).

High spectral resolution lidar (HSRL) (cf. Chapter 5) is the second type of lidar that can be used for the determination of aerosol transmission and light-extinction properties. This lidar relies on the differences in spectral distribution of light elastically backscattered by particles and air molecules. The spectral width of Rayleigh-backscattered photons is increased due to Doppler shifts caused by the thermal motion of the molecules. The thermal motion of aerosol and cloud particles is much slower so their backscatter spectrum remains nearly unchanged. Here, the molecular backscatter channel measures Rayleigh backscattering by blocking the narrow aerosol peak, e.g., by use of an atomic-vapor filter. A second channel may detect the total backscatter or just the central aerosol peak.

Whereas the Rayleigh lidar is operational at day and night, the Raman lidar is mainly used during nighttime, i.e., in the absence of the strong daylight sky background. The strength of Raman signals is a factor of 20 (rotational Raman lines) to 500 (vibration-rotational Raman lines) lower than the one of Rayleigh signals. However, by applying narrow-bandpass filters or a Fabry–Perot interferometer [39] Raman lidar observations are now also possible at daytime with appropriate temporal and spatial resolution [40, 41]. Ground-based solar-blind lidars operating at laser wavelengths well below 300 nm are not appropriate for measurements in the upper troposphere because of strong absorption of laser radiation by ozone.

The determination of the particle extinction coefficient from molecular backscatter signals is rather straightforward. Lidar-ratio assumptions or other critical assumptions are not needed. The advantage of the Raman lidar and the HSRL technique over conventional elastic-backscatter lidar

is already obvious from the respective lidar equation for the molecular backscatter signal,

$$P(R, \lambda_{\text{Ra}}) = \frac{E_0 \eta_{\lambda_{\text{Ra}}}}{R^2} O(R, \lambda_{\text{Ra}}) \beta_{\text{Ra}}(R, \lambda_0) \times \exp \left\{ - \int_0^R [\alpha(r, \lambda_0) + \alpha(r, \lambda_{\text{Ra}})] dr \right\}. \quad (4.13)$$

The coefficient  $\beta_{\text{Ra}}$  denotes Rayleigh backscattering in the HSRL and Raman backscattering in the Raman case. Particle backscattering does not appear in Eq. (4.13). The only particle-scattering effect on the signal strength is attenuation.  $\alpha(R, \lambda_0)$  describes the extinction on the way up to the backscatter region,  $\alpha(R, \lambda_{\text{Ra}})$  the extinction on the way back to the lidar. For the Rayleigh or the rotational Raman case  $\lambda_{\text{Ra}} = \lambda_0$  can be used. However, in the case of a vibration-rotational Raman signal the shift of the wavelength from  $\lambda_0$  before to  $\lambda_{\text{R}}$  after the scattering process must be considered. If, for example, a Nd:YAG laser wavelength of 532 nm is transmitted, the first Stokes vibration-rotation Q branch of nitrogen is centered at  $\lambda_{\text{Ra}} = 607$  nm.

The molecular backscatter coefficient is calculated from the molecular number density  $N_{\text{Ra}}$ , which is the nitrogen or oxygen molecule number density for the Raman case and the air-molecule number density for the Rayleigh case, and the molecular (differential) cross section  $d\sigma_{\text{Ra}}/d\Omega(\pi, \lambda_0)$  for the scattering process (Raman or Rayleigh) at the laser wavelength  $\lambda_0$  and the scattering angle  $\pi$ :

$$\beta_{\text{Ra}}(R, \lambda_0) = N_{\text{Ra}}(R) \frac{d\sigma_{\text{Ra}}}{d\Omega}(\pi, \lambda_0). \quad (4.14)$$

$\beta_{\text{Ra}}(\lambda_0)$  is identical with  $\beta_{\text{mol}}$  in Eq. (4.2), if Eq. (4.13) describes a Rayleigh signal. The molecular number density profile is calculated from actual radiosonde observations or standard-atmosphere temperature and pressure profiles.

After inserting the expressions (4.14) into Eq. (4.13), taking the logarithms of both sides of the resulting equations, differentiating them with respect to  $R$ , and rearranging, we obtain for the total extinction coefficient

$$\alpha(R, \lambda_0) + \alpha(R, \lambda_{\text{Ra}}) = \frac{d}{dR} \ln \frac{N_{\text{Ra}}(R)}{S(R, \lambda_{\text{Ra}})} + \frac{d}{dR} \ln O(R, \lambda_{\text{Ra}}) \quad (4.15)$$

with the range-corrected molecular signals  $S(R, \lambda_{\text{Ra}}) = R^2 P(R, \lambda_{\text{Ra}})$ . The overlap term need not be considered for long distances at which

$O(R, \lambda_{\text{Ra}}) \equiv 1$ . Total laser-beam receiver-field-of-view overlap may in practice not be reached for distances below 2000–3000 m. The measurement range can in these cases be increased (toward the lidar) by correcting for the overlap effect. The correction is based on measurements of the overlap profile with the same lidar under clear sky conditions [42]. However, as can be seen from the relation

$$\frac{d}{dR} \ln O(R, \lambda_{\text{Ra}}) = \frac{1}{O(R, \lambda_{\text{Ra}})} \frac{d}{dR} O(R, \lambda_{\text{Ra}}), \quad (4.16)$$

the determination of the extinction coefficient is rather sensitive to overlap uncertainties close to the lidar where the overlap value is low and changes rapidly with distance.

In the following we concentrate on the optimum measurement range, i.e., we assume proper overlap correction and thus ignore the overlap term in Eq. (4.15). With Eq. (4.3) we can write

$$\begin{aligned} \alpha_{\text{aer}}(R, \lambda_0) + \alpha_{\text{aer}}(R, \lambda_{\text{Ra}}) &= \frac{d}{dR} \ln \frac{N_{\text{Ra}}(R)}{S(R, \lambda_{\text{Ra}})} - \alpha_{\text{mol}}(R, \lambda_0) \\ &\quad - \alpha_{\text{mol}}(R, \lambda_{\text{Ra}}). \end{aligned} \quad (4.17)$$

To obtain the extinction coefficient at the transmitted wavelength we have to introduce the Ångström exponent  $\hat{a}(R)$ , which describes the wavelength dependence of the particle extinction coefficient,

$$\frac{\alpha_{\text{aer}}(\lambda_0)}{\alpha_{\text{aer}}(\lambda_{\text{Ra}})} = \left( \frac{\lambda_{\text{Ra}}}{\lambda_0} \right)^{\hat{a}(R)}, \quad (4.18)$$

(cf. Table 4.1).

Finally we obtain [11]

$$\alpha_{\text{aer}}(R, \lambda_0) = \frac{\frac{d}{dR} \ln \frac{N_{\text{Ra}}(R)}{S(R, \lambda_{\text{Ra}})} - \alpha_{\text{mol}}(R, \lambda_0) - \alpha_{\text{mol}}(R, \lambda_{\text{Ra}})}{1 + \left( \frac{\lambda_0}{\lambda_{\text{Ra}}} \right)^{\hat{a}(R)}}. \quad (4.19)$$

For rotational Raman and HSRL signals the denominator can be set to 2. In contrast to the Klett algorithm, no critical assumption is needed. All the molecular density and scattering terms can be calculated from meteorological or from standard-atmosphere data. Overestimation and underestimation of the  $\hat{a}$  value by 0.5 leads to relative errors of the order of 5%.

As mentioned, in addition to the extinction coefficient, the particle backscatter coefficient can be calculated from the ratio of the aerosol (particle plus Rayleigh) backscatter signal to the molecular backscatter signal as suggested by Cooney et al. [43] and Melfi [44].

The particle backscatter coefficient  $\beta_{\text{aer}}(R, \lambda_0)$ , now explicitly written as a function of the laser wavelength  $\lambda_0$ , can be determined by using both total (particle + molecular) and pure molecular backscatter signals. Two measured signal pairs  $P(R, \lambda_0)$  and  $P(R, \lambda_{\text{Ra}})$  at  $R$  and  $R_0$  are needed. From two lidar signals  $P(R, \lambda_0)$  and  $P(R_0, \lambda_0)$  from total backscatter [Eq. (4.1)] and two more lidar signals  $P(R, \lambda_{\text{Ra}})$  and  $P(R_0, \lambda_{\text{Ra}})$  from molecular backscatter alone [Eq. (4.13)], a solution for the backscatter coefficient  $\beta_{\text{aer}}(R, \lambda_0)$  is obtained by forming the ratio [12]

$$\frac{P(R_0, \lambda_{\text{Ra}})P(R, \lambda_0)}{P(R_0, \lambda_0)P(R, \lambda_{\text{Ra}})}, \quad (4.20)$$

inserting the respective lidar equations for the four signals, and rearranging the resulting equation. The solution is

$$\begin{aligned} & \beta_{\text{aer}}(R, \lambda_0) + \beta_{\text{mol}}(R, \lambda_0) \\ &= [\beta_{\text{aer}}(R_0, \lambda_0) + \beta_{\text{mol}}(R_0, \lambda_0)] \frac{P(R_0, \lambda_{\text{Ra}})P(R, \lambda_0)}{P(R_0, \lambda_0)P(R, \lambda_{\text{Ra}})} \frac{N_{\text{Ra}}(R)}{N_{\text{Ra}}(R_0)} \\ & \quad \times \frac{\exp\{-\int_{R_0}^R [\alpha_{\text{aer}}(r, \lambda_{\text{Ra}}) + \alpha_{\text{mol}}(r, \lambda_{\text{Ra}})] dr\}}{\exp\{-\int_{R_0}^R [\alpha_{\text{aer}}(r, \lambda_0) + \alpha_{\text{mol}}(r, \lambda_0)] dr\}}. \end{aligned} \quad (4.21)$$

If the two signal channels are properly aligned so that  $O(R, \lambda_{\text{Ra}}) = O(R, \lambda_0)$ , then overlap effects cancel out because the backscatter profile is determined from the signal ratio profile  $P(R, \lambda_0)/P(R, \lambda_{\text{Ra}})$ . As a consequence, the backscatter coefficient can be determined even at ranges very close to the lidar, as will be shown in Section 4.4.

As in the Klett procedure, a reference value for particle backscattering at  $R_0$  must be estimated. To reduce the effect of the uncertainty in this estimate on the solution, it is recommended to choose the reference height in the upper troposphere where particle scattering is typically negligible compared to Rayleigh scattering. Then only the air density, the molecular backscattering, and atmospheric extinction properties must be estimated to solve Eq. (4.21). Again, meteorological profiles or standard-atmosphere data are used to calculate air density and molecular backscatter terms. The particle transmission ratio for the height range

between  $R_0$  and  $R$  is estimated from the measured particle extinction profile with the assumption on the wavelength dependence  $\lambda^{\hat{a}}$  as used in Eq. (4.19). In the case of the rotational Raman and HSRL signals spectral transmission corrections are not necessary.

Finally, the height profile of the particle lidar ratio,

$$L_{\text{aer}}(R, \lambda_0) = \frac{\alpha_{\text{aer}}(R, \lambda_0)}{\beta_{\text{aer}}(R, \lambda_0)}, \quad (4.22)$$

is obtained from the profiles of  $\alpha_{\text{aer}}(R, \lambda_0)$  and  $\beta_{\text{aer}}(R, \lambda_0)$  with Eqs. (4.19) and (4.21). For lidars pointed vertically,  $R \equiv z$ .

### 4.3 Retrieval of Physical Properties of Atmospheric Particles

Natural particle size distributions can be described rather well by analytic expressions such as logarithmic-normal distributions (e.g., Hinds [45]):

$$dn(r) = \frac{n_t}{\sqrt{2\pi} \ln \sigma} \exp \left[ -\frac{(\ln r - \ln r_{\text{mod},N})^2}{2 (\ln \sigma)^2} \right] d \ln r. \quad (4.23)$$

$dn(r)$  denotes the number concentration of particles in the radius interval  $[\ln r; \ln r + d \ln r]$ ,  $n_t$  the total number concentration,  $r_{\text{mod},N}$  the mode radius with respect to the number concentration, and  $\sigma$  the mode width, i.e., the geometric standard deviation. Equation (4.23) characterizes a monomodal distribution. Multimodal distributions are sums of  $\geq 2$  monomodal distributions. The surface-area and volume concentrations follow from Eq. (4.23) by multiplication with  $4\pi r^2$  and  $4\pi r^3/3$ , respectively. Other shapes of particle size distributions are found in, e.g., Hinds [45].

The mean and integral properties of the particle ensemble that are calculated from the inverted particle size distribution are the effective radius, i.e., the surface-area-weighted mean radius

$$r_{\text{eff}} = \frac{\int n(r) r^3 dr}{\int n(r) r^2 dr}, \quad (4.24)$$

the total surface-area concentration

$$a_t = 4\pi \int n(r) r^2 dr, \quad (4.25)$$

and the total volume concentration

$$v_t = \frac{4\pi}{3} \int n(r) r^3 dr. \quad (4.26)$$



A variety of methods have been proposed since the early 1970s for the retrieval of microphysical particle properties from lidar measurements. These methods can basically be classified into three distinct groups. The first group deals with the combination of a monostatic lidar with some other instrument, e.g., *in situ* instruments carried aboard an aircraft [46] or balloon [47–50]. Applications are restricted to cases such as field campaigns during which such a combination of instruments on an airplane or other airborne platform is most likely to be at hand. It then provides detailed information on microphysical particle properties [51–53]. Extreme care must be taken, however, in the translation of these data into those amenable to comparison with lidar data. Spatial and temporal proximity of the two instruments is very important in the case of observations of the highly variable tropospheric particles. This constraint is less important for stratospheric particles because of the specific conditions prevailing in the stratosphere (cf. Subsection 4.3.2).

A preferred approach is the combined use of lidar with a sun photometer. In that case the treatment of the data from the two instruments is more straightforward. The latter delivers integrated optical depths of the atmospheric column at multiple wavelengths. A mathematical inversion scheme, which is similar to the methods described in Subsection 4.3.1, is used to derive depth-integrated particle size distributions from the sun photometer observations [54]. The combination with lidar observations then allows a rough estimate of the depth-integrated complex refractive index [17]. A few studies deal with the retrieval of particle size and complex refractive index on the basis of bistatic lidar observations [55–57]. Height profiles of this parameter could be derived with supporting data from additional observations with a monostatic lidar and a sun photometer [58]. Comparison with simultaneous *in situ* observations made aboard an aircraft did not show satisfactory agreement.

The drawback in each case is again that two instruments are needed at the same time and in the same location in order to give reliable data on the same particles. In addition, the lidar and sun photometer point in different directions, i.e., away from the sun and into the sun, respectively. For that reason the constraint of observations of the same ensemble of particles cannot be fulfilled in a strict sense, and thus represents an additional source of error.

In the second class of methods Mie-scattering calculations are intended to reconstruct the optical quantities derived from multiwavelength lidar observations [50, 59–62]. For that purpose parameters such as the shape of the particle size distribution and complex refractive

index are assumed *a priori*. Because of the uncertainties associated with such *a priori* assumptions the application of these methods has been restricted to special cases. In the case of stratospheric particles, i.e., polar stratospheric clouds and ejecta from volcanic eruptions, they were quite successful (cf. Subsection 4.3.2). Investigations of noctilucent clouds in the mesosphere have only recently become possible [63–65]. In the case of tropospheric particles only a crude classification into different types of tropospheric particles is possible [59]. In the special case of desert dust results were unsatisfactory [66].

Finally, the third class is a rigorous mathematical approach on the basis of multiwavelength lidar observations. In that respect the technically robust setup of monostatic Raman lidars is almost exclusively considered. The method uses the spectral information contained in the backscatter and extinction information at multiple wavelengths and its change with particle size. First feasibility studies were made by Uthe [67] and by Uthe et al. [68]. Starting from the work of the early 1980s [57, 69] significant progress has been made. Some exploratory work considered the feasibility of multiwavelength observations with a bistatic lidar [70]. But results are not conclusive for a proper assessment of the potential of this technique in view of the difficulties in connection with the experimental setup. A specific technique deals with the retrieval of particle parameters on the basis of multiple-field-of-view observations in the case of multiple-scattering contributions to aerosol lidar returns [71]. This technique is only applicable for particle size parameters  $>5$ – $10$  [72], and so far only considered the case of single-wavelength lidar. The amount of *a priori* information introduced into the mathematical algorithms can be kept lower compared with the methods belonging to class two. The specific use of mathematical tools makes these techniques very versatile and robust with respect to the highly variable properties of tropospheric particles. The basic properties of the successful algorithms used for the retrieval of microphysical particle properties from multiwavelength lidar sounding will be discussed in the following subsection.

#### 4.3.1 Tropospheric Particles

The method of inversion with regularization with constraints [73] is the standard method for the retrieval of microphysical parameters of tropospheric particles from multiwavelength lidar observations [20, 32, 74–78]. Profiles of the physical particle properties follow from the numerical inversion of the vertically and spectrally resolved particle

backscatter and particle extinction coefficients. The optical data are related to the physical quantities through Fredholm integral equations of the first kind:

$$g_i(\lambda_k) = \int_{r_{\min}}^{r_{\max}} K_i(r, m, \lambda_k, s) v(r) dr + \epsilon_i^{\exp}(\lambda_k),$$

$$i = \beta_{\text{aer}}, \alpha_{\text{aer}}, \quad k = 1, \dots, n. \quad (4.27)$$

The term  $g_i(\lambda_k)$  denotes the optical data at wavelengths  $\lambda_k$  in a specific height  $R$ . For easier reading, reference to height  $R$  will be omitted in the following discussion. The subscript  $i$  denotes the kind of information, i.e., whether it is the particle backscatter ( $\beta_{\text{aer}}$ ) or particle extinction ( $\alpha_{\text{aer}}$ ) coefficient. The data have an error  $\epsilon_i^{\exp}(\lambda_k)$ . The expression  $K_i(r, m, \lambda_k, s)$  describes the kernel efficiencies of backscatter and extinction, respectively. They depend on the radius  $r$  of the particles, their complex refractive index  $m$ , the wavelength  $\lambda_k$  of the interacting light, as well as the shape  $s$  of the particles. For spherical particle geometry the kernel functions  $K_i(r, m, \lambda_k, s)$  are calculated from the respective extinction and backscatter efficiencies  $Q_i(r, m, \lambda_k)$  for individual particles [79] weighted with their geometrical cross section  $\pi r^2$ :

$$K_i(r, m, \lambda_k) = (3/4r) Q_i(r, m, \lambda_k). \quad (4.28)$$

The term  $v(r)$  describes the volume concentration of particles per radius interval  $dr$ . The lower integration limit is defined by  $r_{\min}$ , the radius down to which particles are optically efficient. For measurement wavelengths larger than 355 nm, which is a typical wavelength used for aerosol sounding, the minimum particle size is around 50 nm in radius. The upper limit,  $r_{\max}$ , is the radius at which concentrations are so low that particles no longer contribute significantly to the signal. For typical particle size distributions in the troposphere  $r_{\max}$  is below 10  $\mu\text{m}$ .

In the inversion of Eq. (4.27) the volume concentration is in general preferred over the surface-area or number concentration because it shifts the maximum sensitivity of the kernel efficiencies farther into the optically active range of the investigated particle size distributions. On average this shift leads to a stabilization of the inverse problem (see below). Further improvements of these inversion methods can be expected from a variable use of volume, or surface-area, or number concentration in dependence of the investigated particle size distribution. The main problem which has not been solved yet is how the inversion method by itself can find the most suitable kernel presentation

for each individual data set. First sensitivity studies have been started recently [77, 78].

With the subscript  $p = (i, \lambda_k)$  summarizing the kind and number of optical data, Eq. (4.27) is rewritten into the following form:

$$g_p = \int_{r_{\min}}^{r_{\max}} K_p(r, m) v(r) dr + \epsilon_p^{\exp}. \quad (4.29)$$

Equation (4.29) cannot be solved analytically [80, 81]. The numerical solution process [57, 73] leads to the so-called ill-posed inverse problem [82], which is characterized by the incompleteness of the available information, the non-uniqueness of the solutions, and the non-continuous dependence of the solutions on the input data. Even uncertainties as small as round-off errors in the input data lead to unproportionally large changes in the solution.

The retrieval of microphysical particle properties from lidar measurements belongs to the class of severely ill-posed problems [76]. This definition arises from several features connected to lidar observations. Measurement errors are much larger than round-off errors. Incompleteness is given by the small number of measurement wavelengths, as well as the fact that only backscatter and extinction information is available. The non-uniqueness of the solutions follows from the highly complex structure of tropospheric aerosols. They may be multimodal and of variable shape. The complex refractive index of the particles may be wavelength- or size-dependent, or both. Particle shape often deviates from sphericity. Accordingly different combinations of the target parameters may lead to similar optical spectra within the measurement uncertainty.

The first attempt to derive physical quantities on the basis of lidar observations at multiple wavelengths was undertaken with the so-called randomized-minimization-search technique [69]. This approach did not overcome the instability problem in the solution of Eq. (4.29), when an unknown particle size in combination with an unknown complex refractive index was assumed. The more successful technique of inversion with regularization was introduced after that [57, 83–85]. However, the studies still suffered from unrealistic assumptions on the capabilities of aerosol lidar systems, like a large number of measurement wavelengths and/or features such as the number of extinction channels, or the desired particle information was derived under the assumption of a known complex refractive index. The refinement of this method in combination with the development of powerful aerosol lidar systems [31, 86], which make use of Raman channels, resulted in the determination of particle size

parameters and mean complex refractive indices [74] and thus of the single-scattering albedo [87], which is one of the most important parameters in computer models dealing with climate forcing by aerosols. The most significant step in this development was the reduction of measurement wavelengths to a realistic number, currently provided by modern aerosol lidar instruments.

The three inversion algorithms that are used for the routine retrieval of microphysical properties of tropospheric particles [75–77] make use of the techniques described in the following. Base functions are used to select an appropriate subspace from the solution space. Such projection techniques, which originally were introduced by Backus and Gilbert [88, 89], are a necessary condition to turn the ill-posed inverse problem into a well-posed problem [76]. Only the combination of backscatter and extinction coefficients provides for trustworthy particle parameters [20, 77]. A ratio of two to three for the number of backscatter to extinction coefficients can be considered as the optimum choice for the specifications of an aerosol lidar [77], if simplifications for tropospheric particles like wavelength- and size-independence of the complex refractive index are considered. The three standard wavelengths of a Nd:YAG laser, i.e., 355, 532, and 1064 nm, are the minimum number of wavelengths for particle characterization [76, 77, 90], under the above-mentioned simplifications for the complex refractive index. The accuracy increases if backscatter coefficients at up to six wavelengths are used [20, 57, 77].

For the solution of Eq. (4.29) the investigated size distribution  $v(r)$  is discretized by a linear combination of base functions  $B_j(r)$ , also denoted as B-spline functions, and weight factors  $w_j$ :

$$v(r) = \sum_j w_j B_j(r) + \epsilon^{\text{math}}(r). \quad (4.30)$$

The right-hand side of Eq. (4.30) contains the mathematical residual error  $\epsilon^{\text{math}}(r)$  that is caused by the approximation with base functions. From Eq. (4.30) it is obvious that the inversion codes are not restricted to specific shapes of particle size distributions.

Different shapes of the base functions, denoted as B-splines of order  $l$ , are possible. One has to keep in mind that a good reproduction of natural particle size distributions crucially depends on the shape of the base functions. For this reason histogram columns, denoted as B-spline functions of zero degree [69], are not well suited. Triangle functions, known as B-splines of first degree [75, 77, 85], and parabolic functions,

i.e., B-splines of second degree [91, 92], have shown good performance characteristics. In the latter case the shape already implicitly carries detailed structure of naturally occurring particle size distributions and thus can be considered as a rather constraining approach. In the case discussed in Ben-David et al. [91] the set of base functions was augmented by a special class of Junge-type functions [45] in order to account for such special cases of particle size distributions. A new concept makes use of higher-order polynomials [76], which permit a better reconstruction of highly-structured particle size distributions.

The base functions usually are distributed such that their nodes have the same distance on a non-equidistant scale of particle radii, e.g., on a logarithmic radius scale [75, 77, 85]. In that way the base functions make explicit use of the high dynamic range of particle size distributions which extends over several orders of magnitude. On the other hand, narrow size distributions or distributions consisting of particles around the limit of the optically efficient size range may give better results if their nodes are equally distributed on the linear scale of particle radius.

There is no common recipe for the optimum number of base functions. This number may vary with size range, or number of modes, or particularities of the complex refractive index of the investigated size distribution. Some information may be found in Wolfenbarger and Seinfeld [93], Golub et al. [94], Ben-David et al. [91], Müller et al. [75], and Böckmann [76]. The minimum number of base functions is equal to the number of available data points in the algorithms described in Müller et al. [75] and Veselovskii et al. [77]. Eight base functions provide good resolution of monomodal size distributions and give acceptable estimates for bimodal distributions. Improved results are obtained for higher numbers of base function, i.e., approximately 12–14 base functions. A very high number of 68 base functions was used in theoretical studies in Ben-David et al. [91]. In that case 15 measurement wavelengths, very accurate optical data, and a known complex refractive index were assumed, all of which is rather unrealistic in view of lidar instrumentation. Basically the increase of number of base functions can also be achieved by the use of polynomials of higher order [76]. It has to be observed that an increasing number of base functions again results in increasing destabilization of the inverse problem (e.g., Nychka et al. [95]).

In general the exact position of the investigated particle size distribution along the size range used by Eq. (4.29) is not known. The problem is overcome by the use of a so-called inversion window of variable width

and variable position along the investigated size range [75]. Within this inversion window the base functions are arranged next to each other as described before. No sensible solutions are obtained if the inversion window does not cover the position of the investigated particle size distribution. In that respect the shifting inversion window can be regarded as a filter function. Currently 50 different inversion windows within the particle size range from 0.01 to 10  $\mu\text{m}$  are used to obtain an estimate of the position of the particle size distribution [75, 77]. The polynomials of higher order are restricted to a similar size range [76].

The weight factors follow from inserting Eq. (4.30) into Eq. (4.29), and rewriting it into a vector-matrix equation:

$$\mathbf{g} = \mathbf{A}\mathbf{w} + \boldsymbol{\epsilon}. \quad (4.31)$$

The optical data are written as vector  $\mathbf{g} = [g_p]$ , the weight factors are denoted by  $\mathbf{w} = [w_j]$ , and the errors are described by  $\boldsymbol{\epsilon} = [\epsilon_p]$ .  $\epsilon_p = \epsilon_p^{\text{exp}} + \epsilon_p^{\text{math}}$  is the sum of experimental and mathematical errors. The matrix  $\mathbf{A} = [A_{pj}]$  is called weight matrix [81]. Its elements are given by

$$A_{pj}(m) = \int_{r_{\min}}^{r_{\max}} K_p(r, m) B_j(r) dr. \quad (4.32)$$

The simple solution of Eq. (4.31) for the weight factors,

$$\mathbf{w} = \mathbf{A}^{-1}\mathbf{g} + \boldsymbol{\epsilon}', \quad (4.33)$$

fails to provide reasonable results [81] although the optical data can be reproduced within the error limits  $\boldsymbol{\epsilon}$ . It is explained by the high dynamic range of several orders of magnitude of the elements of  $\mathbf{A}$  and  $\mathbf{A}^{-1}$  [20, 81]. Therefore the term  $\boldsymbol{\epsilon}' = -\mathbf{A}^{-1}\boldsymbol{\epsilon}$ , which describes the respective errors, and  $\mathbf{A}^{-1}$ , which denotes the inverse of the matrix  $\mathbf{A}$ , lead to the aforementioned error amplification and discontinuity of the solutions.

Therefore, a procedure is introduced that we call regularization. This technique selects those solutions for which  $\boldsymbol{\epsilon}$  in Eq. (4.31) drops below a predetermined minimum value  $>0$ . This step is fundamental in the solution process. From first principles it is not possible to exactly reproduce the input optical data from the inversion results. There is always the compromise between the exact reproduction of the optical data and the suppression of error amplification. In the minimization concept, or method of minimum distance [73, 81], the so-called penalty function  $e^2$  is introduced. It is defined via the simple Euclidian norm  $\|\cdot\|$ :

$$e^2 \geq \|\mathbf{A}\mathbf{w} - \mathbf{g}\|^2 + \gamma \Gamma(v). \quad (4.34)$$

The penalty function includes physical constraints that are imposed on the solutions and which are determined by the actual underlying physical problem. One differentiates between descriptive and nondescriptive regularization methods [76]. An example for the first method is the use of *a priori* information about the solution itself, e.g., the exact shape of the particle size distribution. A wrong choice of the shape leads in that case to wrong inversion results. Information on the measurement error is another possibility. The algorithm described here employs the non-descriptive regularization method. In this case “smooth” [75, 80, 81] and positive solutions [75, 96, 97] are assumed for the investigated size distribution. The behavior of the particle size distribution in the vicinity of  $r_{\min}$  and  $r_{\max}$  can also be used as a constraint [75, 77].

The smoothness constraint in Eq. (4.34) is described through the additional penalty term  $\Gamma(v)$ .  $\Gamma(v)$  is a non-negative scalar which measures the deviation of the inverted particle size distribution  $v(r)$  from the requested smoothness. With  $\mathbf{w}^T$  denoting the transposed of the vector  $\mathbf{w}$ , the mathematical definition of smoothness  $\Gamma(v)$  is given by [81]:

$$\Gamma(v) = \mathbf{w}^T \mathbf{H} \mathbf{w}. \quad (4.35)$$

Smoothing is done in terms of the second derivative of the reconstructed particle size distribution. In the case of eight base functions and eight optical data, this matrix is written as:

$$\mathbf{H} = \begin{bmatrix} 1 & -2 & 1 & 0 & 0 & 0 & 0 & 0 \\ -2 & 5 & -4 & 1 & 0 & 0 & 0 & 0 \\ 1 & -4 & 6 & -4 & 1 & 0 & 0 & 0 \\ 0 & 1 & -4 & 6 & -4 & 1 & 0 & 0 \\ 0 & 0 & 1 & -4 & 6 & -4 & 1 & 0 \\ 0 & 0 & 0 & 1 & -4 & 6 & -4 & 1 \\ 0 & 0 & 0 & 0 & 1 & -4 & 5 & -2 \\ 0 & 0 & 0 & 0 & 0 & 1 & -2 & 1 \end{bmatrix}. \quad (4.36)$$

Other forms of smoothing are presented in Twomey [81], but in general are not used in lidar applications. An innovative approach avoids explicit smoothing through the additional penalty term to allow for better retrieval of multimodal size distributions [76].

The solution of the minimization concept follows from writing inequality (4.34) as an equation, and by expressing  $\Gamma(v)$  with Eq. (4.35). With  $\mathbf{T}$  denoting the respective transposed expressions, the weight vector



$\mathbf{w}$  is written as [81]:

$$\mathbf{w} = (\mathbf{A}^T \mathbf{A} + \gamma \mathbf{H})^{-1} \mathbf{A}^T \mathbf{g}. \quad (4.37)$$

The inverse problem is stabilized by the matrix  $(\mathbf{A}^T \mathbf{A} + \gamma \mathbf{H})^{-1} \mathbf{A}^T$ .

Figure 4.1 illustrates the concept of regularization. The influence of  $\mathbf{H}$  is determined by the Lagrange multiplier  $\gamma$ . For  $\gamma = 0$  there is no smoothing, and only the base functions act as regularization (smoothing) parameter [76].  $\gamma \rightarrow \infty$  results in a perfectly smooth solution  $v(r)$  which is, however, independent of  $\mathbf{g}$ . As already explained in the context of inequality (4.34), values of  $0 < \gamma < \infty$  create solutions for which the oscillating behavior is penalized by  $\Gamma(v)$  and thus is suppressed to a certain degree. One chooses as a solution that value of  $\gamma$  for which the complete penalty function in Eq. (4.34) takes a minimum.

The optimum value for the Lagrange multiplier is given by the global minimum of Eq. (4.37), if  $\gamma$  is varied across several orders of magnitude. Different methods can be used to determine this minimum. The exact location of the minimum depends on the respective method, which thus has influence on the quality of the inversion results. A general overview of such methods is found in Engl et al. [98]. With respect to lidar the maximum entropy principle [92], the method of generalized cross-validation [75], the truncated singular value decomposition [76], and the method of minimum discrepancy [77] have been suggested.

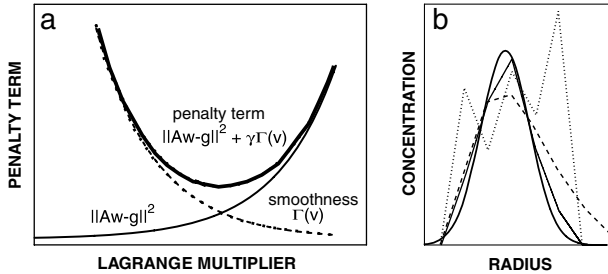


**Fig. 4.1.** (a) Qualitative illustration of the reconstruction error for increasing smoothing. Shown is the penalty term (thick solid), the error of the reconstructed optical data (thin solid), and the error caused by the smoothing term (dashed). (b) Qualitative effect on the accuracy of the reconstructed particle size distribution. Shown is the case with insufficient smoothing which leads to oscillating solutions (dotted), ideal smoothing, which leads to an ideal reconstruction (thin solid), and the case of strong smoothing which again leads to a false reconstruction (dashed). Also shown is the theoretical particle size distribution (thick solid).

Generalized cross-validation (GCV) has been shown to work quite well [20]. The method explicitly uses the relationship among the input data. It neither needs an *a priori* estimation of the expected error in the data nor an *a priori* assumption of the solution or the statistical and systematic errors. There is only little tendency toward oversmoothing, and low sensitivity to statistical and systematic errors. The closed expression for the calculation of the GCV parameter  $P_{\text{GCV}}$  is written as

$$P_{\text{GCV}}(\gamma) = \frac{\frac{1}{p} \|\mathbf{I} - \mathbf{M}(\gamma)\| \mathbf{g}\|^2}{\left\{ \frac{1}{p} \text{trace} [\mathbf{I} - \mathbf{M}(\gamma)] \right\}^2} \longrightarrow \min. \quad (4.38)$$

$\mathbf{I}$  is the unit matrix. The influence matrix

$$\mathbf{M}(\gamma) = \mathbf{A} [\mathbf{A}^T \mathbf{A} + \gamma \mathbf{H}]^{-1} \mathbf{A}^T \quad (4.39)$$

is the product of the kernel matrix  $\mathbf{A}$  and the matrix  $[\mathbf{A}^T \mathbf{A} + \gamma \mathbf{H}]^{-1} \mathbf{A}^T$  of Eq. (4.37) [99].

A modified form of the original minimum discrepancy principle also showed good performance [77]. The need for knowing *a priori* the measurement error has been overcome by the use of the modulus  $|v(r)|$  of the retrieved particle size distribution. In that case the minimum deviation is calculated from the difference between input optical data and the optical data obtained from  $|v(r)|$ . The parameter of the modified minimum discrepancy  $P_{\text{MMD}}$  is calculated as

$$P_{\text{MMD}}(\gamma) = \frac{1}{p} \frac{\|\mathbf{g} - \mathbf{A}|\mathbf{v}|\|}{\mathbf{g}} \longrightarrow \min. \quad (4.40)$$

The global minimum of regularization curves may become rather shallow under certain circumstances [20]. In that case the solutions are not well defined. Averaging of many solutions across the shallow minimum of the discrepancy curve is an elegant new concept [77]. It significantly improves the inversion results and even provides a new approach to error estimation in the inversion.

The discussion given to this point has described the retrieval of the particle volume concentrations for one complex refractive index and for one input optical data set for the (50) different inversion windows. In the case of regularization with GCV this solution space is further constrained. If the measurement errors are known, only those particle volume concentrations are accepted for which the recalculated optical

data are within the uncertainty of the input optical data. In the case of the minimum discrepancy principle the additional constraintment of the solution space implicitly follows from the regularization step.

The solution process described above now has to be carried out for a range of wavelength- and size-independent complex refractive indices, which are given by the kernel functions in Eq. (4.29) [20]. For that purpose one chooses a grid of values for the complex refractive index. The imposed constraints do not allow solutions for some of the complex refractive indices. Consequently, the solution space of this parameter is determined. However, the solution space may be unacceptably large. In such cases the solution space may be further constrained. For example solutions that have optical data closest to the input optical data may serve as constraint. Again it must be observed that the exact reproduction of the input optical data is not possible for theoretical reasons. In addition one can make use of the statistical information within the solution space. If the complex refractive index is correctly chosen, there may be many more inversion windows that provide for sensible particle size distributions compared with complex refractive indices far away from the correct value. However, this property cannot be considered as a general rule. If such solution spaces are visualized in terms of a matrix-like scheme, with  $x$  and  $y$  representing the real and imaginary axes, one obtains solutions along a diagonal. At some point within this diagonal many more solutions are found than in another region, or the reconstructed optical data, which belong to these solutions, may exhibit a lower deviation to the input optical data compared with the conditions in neighboring areas of the matrix. This area of decreased reconstruction error may serve as a further constrained solution space. Examples for such matrices are found in Müller et al. [87, 90] and Böckmann [76].

Solutions have to be determined for the input data varied within their measurement uncertainty. It is not clear how many variations must be performed until statistically significant results are obtained. This uncertainty is again caused by the numerical solution of the ill-posed problem, which is highly nonlinear, and the actual physical properties that underly the optical data. A discussion can be found in Müller et al. [20] and Böckmann [76]. It is assumed that in general 10–20 different runs provide reliable results.

A severe problem in the inversion is a trustworthy error analysis. Because of the numerical solution process and the highly nonlinear behavior of Eq. (4.29) standard techniques of determining error propagation fail. The methods described above have shown to give acceptable error estimates [32, 77], but cannot be considered as the final solution to

this problem. Future work will also focus on resolving the simplifications that have to be made so far and which present additional sources of uncertainty. Table 4.1 lists the single-scattering albedo which is a wavelength dependent parameter. This wavelength dependence is the result of particle size as well as the fact that particle size distributions may possess a wavelength- and/or size-dependent complex refractive index. The investigation of how accurately parameters of particles of non-spherical geometry can be retrieved will be another challenge. For that purpose particle depolarization measurements at one or several wavelengths will have to be considered in the data retrieval. The most important example of this class of particles is desert dust, see Table 4.1.

### 4.3.2 Stratospheric Particles

Three different approaches have been applied to describe the long-term trends in the microphysical properties of the stratospheric aerosol layer after the Mount Pinatubo eruption: The conventional, single-wavelength-lidar technique [48, 100, 101], the multiwavelength-backscatter-lidar technique [50, 60–62, 102, 103], and the Raman-lidar technique [36, 49]. Most simple and reliable are the conventional and the Raman lidar technique. These two methods were compared in terms of surface-area and mass concentrations based on dense, 5-year Pinatubo data sets [104].

The conventional and the Raman lidar technique make use of the following relationship between the total surface-area concentration  $a_t(z)$  and total volume concentration  $v_t(z)$ , and the backscatter and extinction coefficients at height  $z$  at a single wavelength [49]:

$$\frac{a_t(z)}{\beta_{\text{aer}}(z)} = \frac{4}{Q_{\beta, \text{eff}}(z)}, \quad (4.41)$$

$$\frac{v_t(z)}{\beta_{\text{aer}}(z)} = \frac{4r_{\text{eff}}(z)}{3Q_{\beta, \text{eff}}(z)}, \quad (4.42)$$

$$\frac{a_t(z)}{\alpha_{\text{aer}}(z)} = \frac{4}{Q_{\alpha, \text{eff}}(z)}, \quad (4.43)$$

$$\frac{v_t(z)}{\alpha_{\text{aer}}(z)} = \frac{4r_{\text{eff}}(z)}{3Q_{\alpha, \text{eff}}(z)}, \quad (4.44)$$

with the effective scattering efficiencies  $Q_{\alpha, \text{eff}}(z)$  and  $Q_{\beta, \text{eff}}(z)$

$$Q_{\alpha, \beta, \text{eff}}(m, \lambda, z) = \frac{\int_0^\infty Q_{\alpha, \beta}(r, m, \lambda) n(r, z) r^2 dr}{\int_0^\infty n(r, z) r^2 dr}. \quad (4.45)$$

The  $Q_{\alpha,\beta}$  [cf. Eq. (4.28)] and, thus,  $Q_{\alpha,\beta,\text{eff}}(z)$  depend on the particle size distribution  $n(r, z)$  and the refractive index  $m$  of the aerosol particles which, in the case of volcanic aerosol, consist of sulfuric acid and water. The sulfuric-acid content of the droplets is mainly a function of temperature and varies between 40% and 85% for temperatures between 195 and 250 K. The refractive index can thus be accurately determined by means of temperature profiles routinely measured with radiosondes. The size distribution  $n(z)$  of stratospheric particles is well described by a bimodal logarithmic-normal distribution. Typical median (mode) radii are 0.07–0.1  $\mu\text{m}$  for the stratospheric background mode and 0.3–0.6  $\mu\text{m}$  for the volcanic mode [48].

The surface-area and volume concentrations,  $a_t(z)$  and  $v_t(z)$ , are finally determined by multiplying the backscatter coefficients and/or the extinction coefficients with the respective conversion factors [Eqs. (4.41)–(4.44)]. The mass concentration is obtained by multiplying the volume concentration  $v_t(z)$  with the specific gravity of the sulfuric acid droplets. Finally, the effective radius as defined by Eq. (4.24) is simply given by  $3v_t/a_t$ .

As discussed and illustrated by Jäger and Hofmann [48] and by Jäger and Deshler [101] who performed extensive Mie-scattering calculations and evaluated in this way 20-year measurements of the aerosol size distribution with balloon-borne optical particle counters at Laramie, Wyoming, the conversion factors change considerably with time and height during the first three years after a major volcanic eruption. This change is caused by the change of the particle spectrum, especially by the removal of the second, volcanically induced large particle mode as a result of size-dependent gravitational settling. In that case the effective radius and the scattering efficiencies of the scatterers decrease. It was found that the seasonally averaged conversion factors in the lower stratosphere dropped by a factor of 1.5–3 in the first winter after the El Chichón and the Pinatubo eruptions compared to the respective pre-eruption values. They slowly returned to stratospheric background values during the following three years. Thus, to obtain reliable results from the conventional backscatter-lidar measurements, a time- and height-dependent stratospheric aerosol model is used today that is based on the Laramie measurements. By means of this model, seasonally averaged conversion factors for several stratospheric layers are determined and applied to the lidar data. As a consequence of this procedure, the backscatter-lidar technique is restricted to midlatitudes and cannot be used in the tropics or in polar regions because the temporal and vertical behavior of the aerosol characteristics are unknown there. Furthermore, it is assumed

that the aerosol characteristics found above Laramie at  $41^\circ \text{N}$  for a certain height level and time interval are equal to the microphysical properties in the stratosphere over Garmisch-Partenkirchen ( $47.5^\circ \text{N}$ ) in the same height region and for the same time period. Hence, the model-derived conversion factors may be useful in the study of the long-term trend of volcanic effects based on monthly or seasonal mean values of particle parameters, but care must be taken in the interpretation of individual observations of height profiles of the surface-area and volume or mass concentrations.

As shown by Wandinger et al. [49], the conversion factors are functions of the extinction-to-backscatter ratio. The factors can therefore directly be determined with sufficient accuracy from the measured lidar ratio in the case of Raman-lidar observations so that, in the second step, the microphysical properties can directly be determined from the backscatter and extinction profiles. No aerosol model is necessary. This was found from calculations of conversion factors for a large number of measured stratospheric particle size distributions. The Raman-lidar method can therefore be used at very different places on the globe. It was finally shown that the two-wavelength Raman-lidar technique at laser wavelengths of 355 and 532 nm is most promising for stratospheric aerosol profiling. The shorter wavelength is more sensitive to changes in the optical characteristics and in the conversion factors several years after the eruption when the background mode dominates the optical effects. The longer wavelength is optimum during the first two years after the eruption when the volcanic aerosol mode determines the conversion factors. The principal-component analysis of multiwavelength Raman lidar observations as suggested by Donovan and Carswell [35] may be regarded as an alternative approach to the technique suggested by Wandinger et al. [49].

Multiwavelength backscatter lidar is another promising technique for the determination of microphysical parameters. Again, a time- and height-dependent aerosol model is not required. However, this technique suffers from the fact that the key information used in the retrieval procedure is the spectral slope of the backscatter coefficient determined from elastic backscatter signals at two to four different wavelengths. As outlined in Section 4.2, the signal profiles must be calibrated which is crucial for wavelengths  $\geq 1 \mu\text{m}$ . In addition, profiles of the lidar ratio at the laser wavelengths have to be estimated. The assumed lidar ratio profiles sensitively affect the particle backscatter determination at wavelengths  $\leq 532 \text{ nm}$ . As a consequence, the spectral slope of the backscatter coefficient can only roughly be estimated.

Different approaches to retrieve the physical properties can be found in the literature. The basic idea is the comparison of the spectral slope and strength of the backscatter coefficient obtained from Mie-scattering calculations as a function of number concentration, mode radius, and width of monomodal logarithmic-normal size distributions with the observed backscatter-coefficient spectrum. From the size distribution the parameters can then be calculated that best explain the lidar observations of surface-area, volume, and mass concentrations and the effective radius.

## 4.4 Measurement Examples

### 4.4.1 Indo-Asian Haze Over the Tropical Indian Ocean

Figure 4.2 shows a lidar measurement taken during the Indian Ocean Experiment (INDOEX), which was conducted in February and March of 1999 in the tropical Indian Ocean. The observation was made with a scanning 6-wavelength-11-channel aerosol lidar [86] at Hulule Island, Maldives ( $4^{\circ}$  N,  $73^{\circ}$  E). Two Nd:YAG and two dye lasers served as radiation sources at 355, 400, 532, 710, 800, and 1064 nm. A beam combination unit was used to align all six laser beams onto one optical axis. The combined beam was then directed into the atmosphere at any zenith angle between  $-90^{\circ}$  and  $+90^{\circ}$  by means of a steerable mirror.

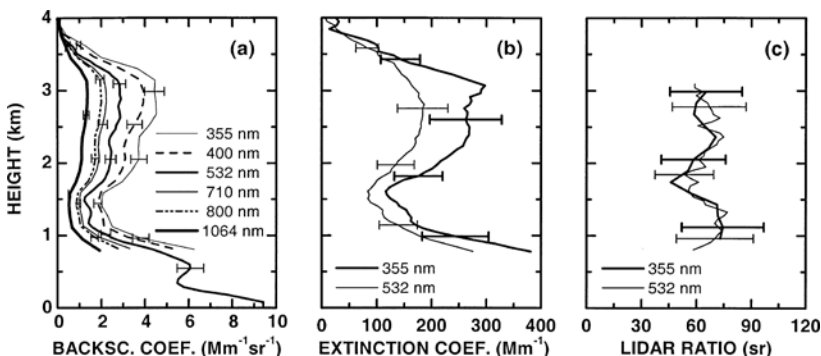


**Fig. 4.2.** Profiles of (a) backscatter and (b) extinction coefficient, and (c) lidar ratio measured on March 25, 1999 [105]. Error bars denote standard deviations caused by signal noise and systematic errors resulting from the estimates of input parameters. Because of large uncertainties introduced by the overlap effect and detector problems at 355 nm only the 532-nm backscatter profile is trustworthy down to the ground.

The profiles shown in Fig. 4.2 were taken at a zenith angle of  $30^\circ$ . In addition to the return signals elastically backscattered by air molecules and particles at the laser wavelengths, Raman signals from nitrogen molecules were detected at 387 nm (355 nm primary wavelength) and 607 nm (532 nm primary wavelength), and from water-vapor molecules at 660 nm (532 nm primary wavelength). At 710 nm, the cross- and parallel-polarized component (with respect to the plane of polarization of the emitted laser light) of the backscattered signals were detected separately.

The example in Fig. 4.2 shows strongly absorbing anthropogenic pollution advected from the Indian subcontinent to the lidar site [105]. The particle backscatter and extinction coefficients at 355 and 532 nm were determined using the Raman-lidar method (Subsection 4.2.2). The backscatter profiles at 400, 710, 800, and 1064 nm were obtained with the Klett method (Subsection 4.2.1). The lidar ratio profiles were calculated from the backscatter and extinction profiles at 355 and 532 nm. Detector problems at 355 nm and large uncertainties in the overlap correction prohibited the computation of the optical parameters below about 800 m height.

As can be seen in Fig. 4.2, several aerosol layers were present over the Maldives on March 25, 1999. Above the polluted marine boundary layer that reached into heights of 1000 m, a continental Indian pollution plume extended up to 4000 m height. The extinction values were rather large with values of 150–300  $\text{Mm}^{-1}$  ( $\text{Mm}^{-1} = 10^{-6} \text{ m}^{-1}$ ) in the lofted, free tropospheric aerosol layer. The 532-nm particle optical depth was close to 0.6. The lofted layer contributed more than 60% to the total particle optical depth. The optical depth of the marine boundary layer was 0.2 and a factor of 2–3 larger than values obtained under unperturbed, clean conditions. The lidar ratios were often between 60 and 90 sr during INDOEX [30]. This finding is consistent with the presence of a considerable amount of strongly absorbing particles in South Asian aerosol pollution.

Such an aerosol layering as presented in Fig. 4.2 cannot be resolved with ground-based or spaceborne passive remote sensing. Only active remote sensing allows a detailed, height-resolved analysis of this interesting and, from the point of view of climate and environmental research, very important measurement case.

Figure 4.3 shows the corresponding profiles of the microphysical properties determined with the inversion scheme outlined in Subsection 4.3.1. Mean effective radii are approximately  $0.17 \mu\text{m}$  below 1000 m height. Rather height-independent mean values of  $0.14\text{--}0.18 \mu\text{m}$  in



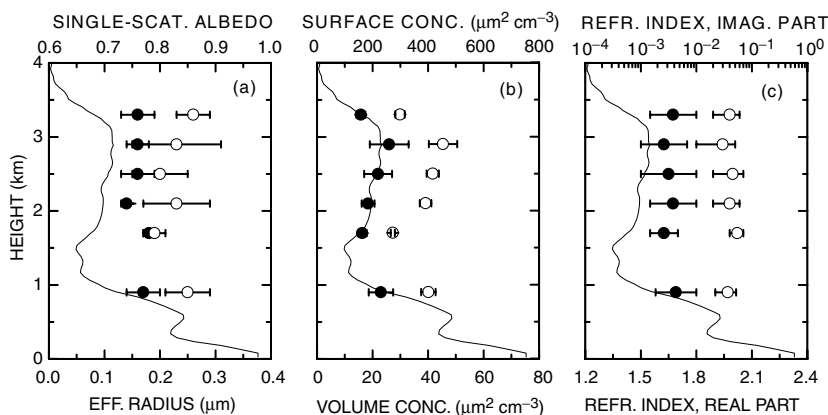


**Fig. 4.3.** Profiles of (a) effective radius (●) and single-scattering albedo (○), (b) volume concentration (●) and surface-area concentration (○), and (c) mean values of the real (●) and imaginary part (○) of the complex refractive index [106, 107]. The error bars for the particle size parameters indicate the standard deviation. For the inversion the profiles were averaged across layers of 400 m thickness. The solid curve in each of the figures shows the 532-nm particle backscatter coefficient.

the upper layer indicate a well-mixed pollution plume of aged, anthropogenic aerosol particles. The air masses traveled about six days from the highly industrialized and populated northern parts of India. They crossed the Bay of Bengal before they reached the Maldives with the prevailing northeast trade winds. The single-scattering albedo ranged between 0.79 and 0.86. These low values are also representative for strongly absorbing particles which are believed to lead to a warming of the climate, as opposed to sulfate particles which are the main component of North American and European aerosols. The imaginary part shows rather high values between  $0.01i$  and  $0.08i$  and is much larger than the typical value found for marine aerosols. The real part of the refractive index varies between 1.5 and 1.8, with values centered around 1.65. The mean volume concentration varies between  $16$  and  $26 \mu\text{m}^3/\text{cm}^3$ . Mean surface-area concentrations vary between  $270$  and  $450 \mu\text{m}^2/\text{cm}^3$ .

In this example of an Indian pollution plume, the imaginary part of the complex refractive index is an order of magnitude larger than corresponding values found for non-absorbing European pollution observed during the Aerosol Characterization Experiment 2 [108]. The volume concentration is approximately 40% larger, the surface-area concentration is 10–20% larger. Particles from biomass burning observed over Germany after long-range transport from northwest Canada during the Lindenberg

Aerosol Characterization Experiment showed comparably large imaginary parts [32]. However, volume and surface-area concentrations were on average lower by a factor of 2–10.

#### 4.4.2 Mount–Pinatubo Aerosol Layer

Figures 4.4 and 4.5 show two examples of stratospheric aerosol observations over Germany after the Pinatubo eruption which is believed to be the strongest perturbation of the stratospheric aerosol layer in the past century. The Raman lidar measurement in Fig. 4.4 was performed at Geesthacht (GKSS Research Center, 53.4° N, 10.4° E) during the first spring after the eruption when the aerosol loading was highest. Extinction coefficients reached values characteristic for thin cirrus and the surface-area and mass concentrations were a factor of 30–100 above the stratospheric background values. The surface-area concentration was clearly above the threshold value of 15–20 mm<sup>2</sup>m<sup>−3</sup> (1 mm<sup>2</sup>m<sup>−3</sup> = 1 μm<sup>2</sup>cm<sup>−3</sup>) during the first winters after the Pinatubo eruption. At such high values of surface area available for heterogeneous chemical reactions a significant ozone reduction was observed in many places around the globe. The uncertainties in the physical parameters retrieved with the Raman-lidar method (Subsection 4.3.2) are discussed in detail by Wandinger et al. [49]. The error bars in Fig. 4.4 include both statistical and systematic (retrieval) errors. The uncertainties are relatively large because of the ozone absorption correction necessary at the measurement wavelength of 308 nm. The overall uncertainties would decrease by about 30% in the case of a laser wavelength of 355 nm at which absorption by ozone is negligible.

The evolution of the stratospheric aerosol layer in terms of the mass concentration over Geesthacht in northern Germany and Garmisch-Partenkirchen (47.5° N, 11.1° E) in southern Germany is presented in Fig. 4.5. Monthly mean values obtained with the conventional lidar and the Raman-lidar technique are compared. A very good agreement was found in the central and lower part of the volcanic aerosol layer. The perturbation of the stratospheric aerosol layer declined with a 1/e decay time of 13 to 13.5 months in terms of the mass concentration.

The good agreement between the two data sets confirms, on the one hand, the capability of a conventional backscatter lidar, constrained to a realistic aerosol model, to monitor the aerosol parameters most important for climate and ozone–chemistry research. These measurements were carried out to yield monthly or seasonal mean values. The agreement

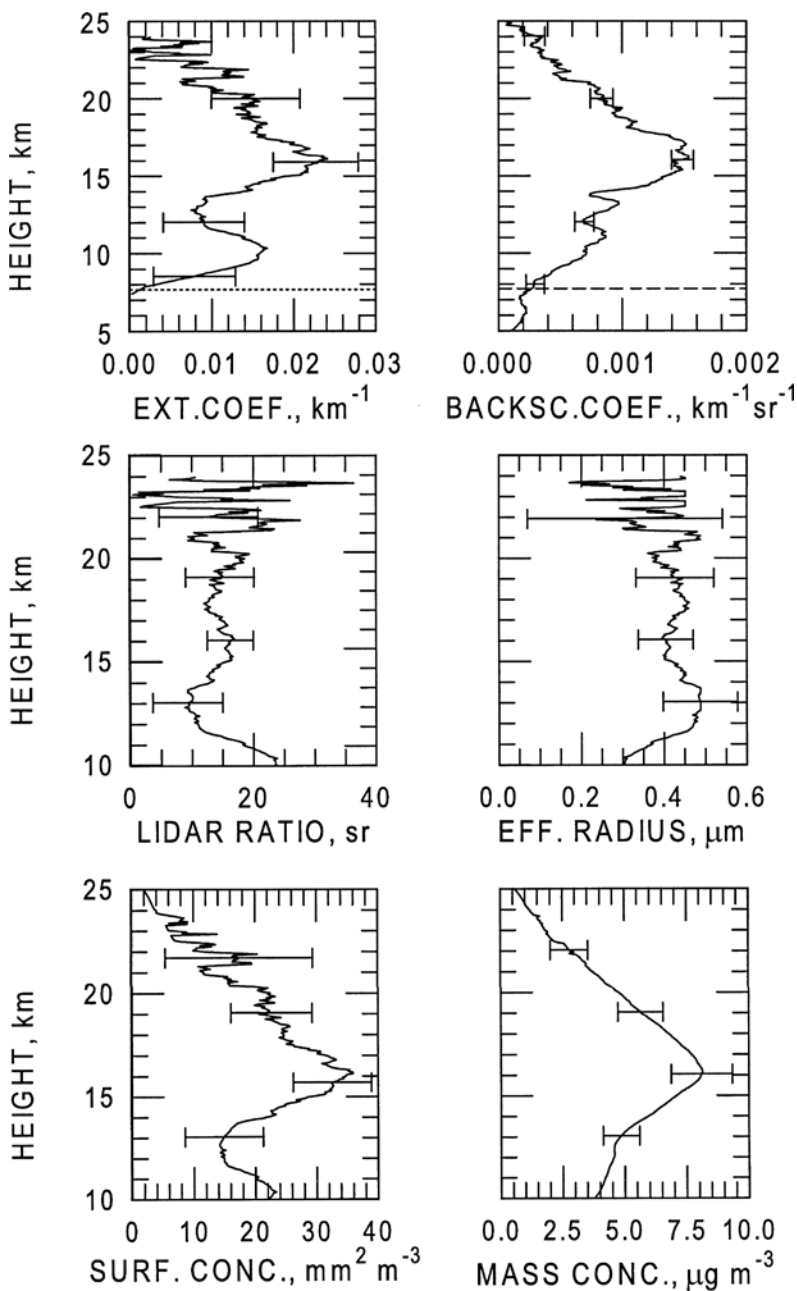


**Fig. 4.4.** Particle extinction and backscatter coefficients at 308 nm, the extinction-to-backscatter ratio at 308 nm, the effective radius of the particle size distribution, and the particle surface-area and mass concentrations [36]. The measurement was taken on April 4, 1992. The dashed lines indicate the tropopause. The optical depth of the stratospheric aerosol layer was 0.25. Error bars indicate the overall retrieval error.

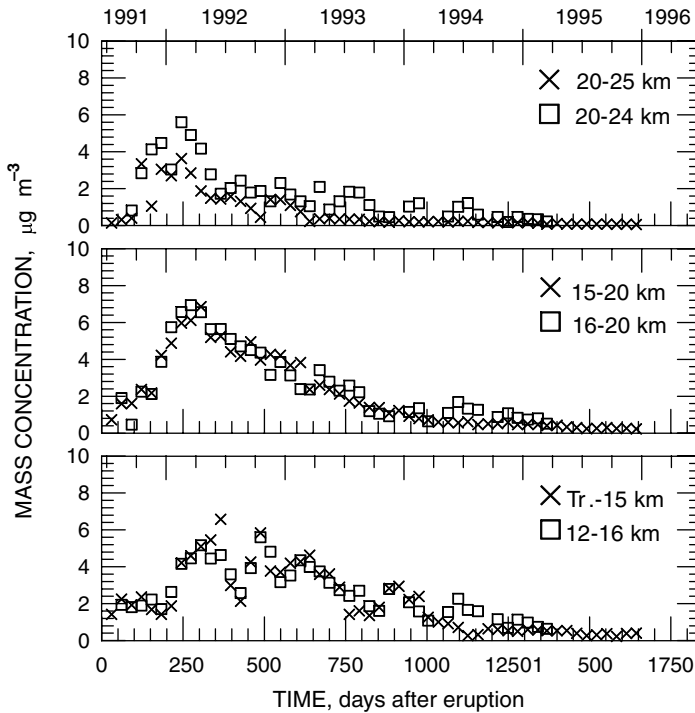


**Fig. 4.5.** Monthly mean layer-averaged mass concentrations derived from lidar observations at Geesthacht (squares, layers from 12–16 km, 16–20 km, and 20–24 km height) and at Garmisch-Partenkirchen (crosses, layers from the tropopause to 15 km height, 15–20 km, and 20–25 km height) [104]. In terms of the monthly mean mass values integrated from the tropopause to 25 km height, the atmospheric perturbation declined with a  $1/e$ -folding decay time of 13.5 months (Garmisch-Partenkirchen) and 13.3 months (Geesthacht).

corroborates the reliability not only of the Pinatubo-related findings but also of the El-Chichón observations which had already been performed in the way described here. This technique can thus be applied to many single-wavelength lidars monitoring the stratosphere both at northern and southern midlatitudes. On the other hand, the comparison impressively demonstrated the usefulness and, because of its longer list of accessible aerosol parameters, superiority of an aerosol Raman lidar. Because of the attractiveness of this technique, Raman channels are being or have been implemented in many lidars around the world during the past years. It remains to mention that today's aerosol lidar technology enables us to provide the scientific community with vertically resolved information about the relevant aerosol properties needed to

properly describe the impact of aerosols on the Earth's radiation budget and atmospheric chemical processes.

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## High Spectral Resolution Lidar

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### 5.1 Introduction

Standard lidar systems provide profiles of the attenuated backscatter signal. These can be assembled into images with spatial and temporal resolution adequate for practically all problems of atmospheric research. However, the attenuated backscatter signal is a quantity that is composed of different atmospheric properties. Extremely valuable for an assessment of the state of the atmosphere, they are hard to extract from the measured profiles of the attenuated backscatter signal.

We start from the familiar lidar equation

$$P(r) = P_0 \eta \left( \frac{A}{r^2} \right) O(r) \left( \frac{ct}{2} \right) \beta(r) \exp \left[ -2 \int_0^r \alpha(r') dr' \right] \quad (5.1)$$

in which  $P(r)$  is the power received from range  $r$ ,  $P_0$  is the average transmitted power during the laser pulse,  $\eta$  is the receiver efficiency,  $A$  is the receiver area,  $r$  is the range to the scattering volume,  $O(r)$  is the laser-beam receiver-field-of-view overlap function,  $c$  is the speed of light,  $t$  is the laser pulse duration, and  $\beta$  and  $\alpha$  are the atmospheric backscatter coefficient and atmospheric extinction coefficient at range  $r$  (see Chapter 1). The integrated extinction coefficient

$$\tau(0, r) = \int_0^r \alpha(r) dr \quad (5.1a)$$

or, more generally,

$$\tau(r_1, r_2) = \int_{r_1}^{r_2} \alpha(r) dr \quad (5.2)$$

is known as the optical depth or optical thickness of the atmosphere from the ground to height  $r$  or of a layer between  $r_1$  and  $r_2$ , respectively.

Equation (5.1) relates profiles of one measured quantity with the profiles of the unknown atmospheric backscatter coefficient  $\beta$  and the unknown atmospheric extinction coefficient  $\alpha$ . If we consider that each of these is the sum of a molecular and an aerosol component and that extinction, or the removal of light from a straight path through the atmosphere, is caused by both the part that is scattered and the part that is absorbed, or

$$\beta = \beta_{\text{mol}} + \beta_{\text{aer}} \quad \text{and} \quad (5.3)$$

$$\alpha = \alpha_{\text{mol},\text{sca}} + \alpha_{\text{mol},\text{abs}} + \alpha_{\text{aer},\text{sca}} + \alpha_{\text{aer},\text{abs}}, \quad (5.4)$$

then we have to solve Eq. (5.1) at each height for six unknowns, a task that is clearly impossible to carry out. Our knowledge of the atmosphere allows us to reduce the six primary unknowns to two by a procedure that, although well known, shall be briefly recalled here.

The magnitude of  $\beta_{\text{mol}}$  is obtained from Rayleigh scattering theory [1, 2].  $\beta_{\text{mol}}$  obeys, for all practical purposes, a proportionality with atmospheric density. So if ground-level atmospheric temperature and pressure and the shape of one of the profiles (usually the temperature profile, from a radiosonde ascent or, better, a lidar) are known, then the profile of  $\beta_{\text{mol}}$  is also known. The proportionality factor is

$$\frac{\beta_{\text{mol}}(r)}{\beta_{\text{mol}}^{\text{STP}}} = 1.47 \times 10^{-6} \times \left( \frac{550 \text{ nm}}{\lambda} \right)^4 \text{ m}^{-1} \text{ sr}^{-1}, \quad (5.5)$$

with standard temperature and pressure (STP) defined as 0°C and 1013.25 hPa, conditions at which the atmosphere contains  $2.69 \times 10^{25}$  molecules per  $\text{m}^3$  [3].  $\beta_{\text{aer}}$  is also known as the absolute (nonnormalized) aerosol scattering phase function at scattering angle  $180^\circ$  or  $\pi$ ,  $\wp(\pi)$ .

$\alpha_{\text{mol},\text{sca}}$  is strictly proportional to  $\beta_{\text{mol}}(r)$  and is given by

$$\alpha_{\text{mol},\text{sca}} = \beta_{\text{mol}}(r) \times (8/3)\pi \text{ sr}. \quad (5.6)$$

$\alpha_{\text{mol},\text{abs}}$  is simply assumed to be zero. Clearly, this is not justified when, e.g., sizable concentrations of ozone are present and lidar wavelengths

are used at which ozone is known to absorb. Special techniques must then be used, e.g., correction algorithms based on the known concentration profiles measured with a lidar (see Chapter 7).  $\beta_{\text{aer}}$ ,  $\alpha_{\text{aer,sca}}$ , and  $\alpha_{\text{aer,abs}}$  are related with one another in a way determined by particle size, shape, and the real and imaginary part of the refractive index—provided the particles are homogeneous, which is not always the case. Clearly this relationship requires too much information and is too complicated to be useful for practical purposes. Therefore,  $\alpha_{\text{aer,sca}}$  and  $\alpha_{\text{aer,abs}}$  are considered together as one unknown  $\alpha_{\text{aer}} = \alpha_{\text{aer,sca}} + \alpha_{\text{aer,abs}}$ , and  $\beta_{\text{aer}}$  as the other. This still leaves us with two unknown profiles to be determined from one profile of measured data.

Essentially two methods have been in use to solve the problem. One, known as the Klett method, is the assumption of a functional dependency of  $\alpha_{\text{aer}}$  and  $\beta_{\text{aer}}$ , which then reduces the number of unknowns to one. This method is described and discussed in detail in Chapter 4; it suffers from the great variability of aerosols, which leads to deviations from the proportionality of  $\alpha_{\text{aer}}$  and  $\beta_{\text{aer}}$  vital for the Klett method to work. Another is the measurement of two lidar profiles in one of which  $\beta_{\text{aer}} = 0$ . This is the case in Raman lidar. Only molecules, not aerosols, contribute to the inelastic, i.e., frequency-shifted, Raman backscatter profile produced by molecular nitrogen and oxygen. From the fact that the elastic lidar return signal is affected by both  $\alpha_{\text{aer}}$  and  $\beta_{\text{aer}}$ , but the Raman lidar return by  $\alpha_{\text{aer}}$  alone, the two profiles can be solved for the two unknowns (Chapter 4). However, Raman scattering is weak; less than one photon will be scattered into one of the vibrational Raman lines for each thousand photons that are elastically scattered by a molecule. Furthermore, the spectrum of the scattered photons is broad, reflecting the large number of allowed rotational transitions of the scattering molecule. Raman lidars require powerful lasers and large telescopes in order to provide sufficient signal strength. Daytime operation is difficult because the small Raman signal must compete with scattered sunlight in the optical bandwidth required to collect the rotational lines of the Raman signal.

## 5.2 The Principle of High Spectral Resolution Lidar

Another idea based on the use of two measured profiles instead of just one is high spectral resolution lidar, or HSRL. This method utilizes the Doppler frequency shifts produced when photons are scattered

from molecules in random thermal motion. The Maxwellian distribution of molecular velocities has a width of  $\sim 300$  m/s that produces Doppler shifts of  $\sim 1$  GHz. In contrast, aerosols, cloud particles, and other particulate matter move with velocities determined by the wind ( $\sim 10$  m/s) and turbulence ( $\sim 1$  m/s) producing Doppler shifts of  $\sim 30$  MHz and  $\sim 3$  MHz, respectively. As a result, the frequency distribution of light backscattered from the atmosphere consists of a narrow spike near the frequency of the laser transmitter caused by particulate scattering riding on a much broader distribution produced by molecular scattering (see Fig. 5.1).

As in Raman lidar, there are two lidar equations instead of just one, but the structure of the equations is a lot more similar as only one wavelength is involved. Dropping the wavelength dependence, we have for

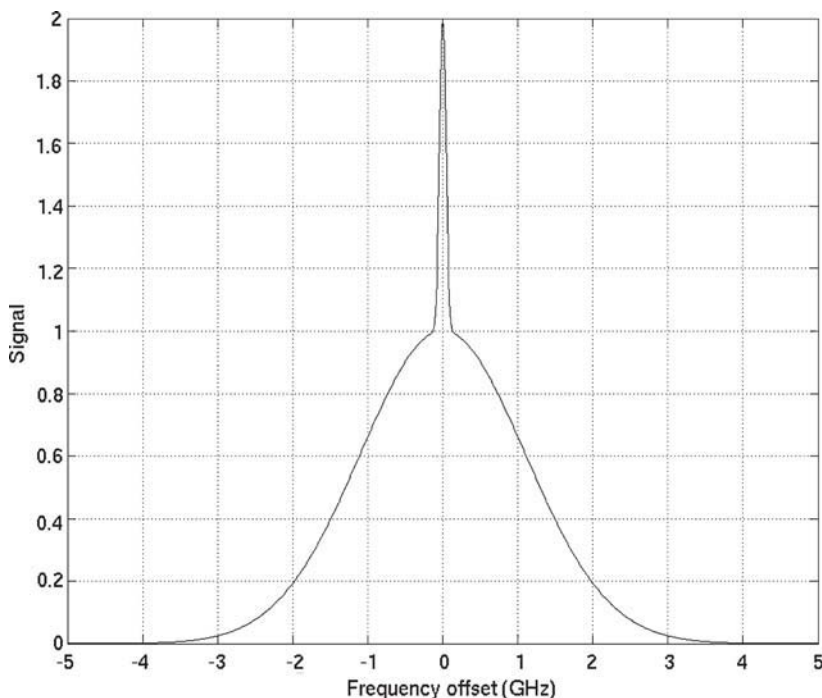


**Fig. 5.1.** Spectral profile of backscattering from a mixture of molecules and aerosols for a temperature of 300 K. The spectral width of the narrow aerosol return is normally determined by the line width of the transmitting laser.

the molecule and the aerosol signal

$$P_{\text{mol}}(r) = K_{\text{mol}} r^{-2} O(r) \beta_{\text{mol}}(r) \exp \left( -2 \int_0^r \alpha(r') dr' \right) \text{ and} \quad (5.7)$$

$$P_{\text{aer}}(r) = K_{\text{aer}} r^{-2} O(r) \beta_{\text{aer}}(r) \exp \left( -2 \int_0^r \alpha(r') dr' \right) \quad (5.8)$$

in which the terms  $K_{\text{mol}}$  and  $K_{\text{aer}}$  contain all range-independent variables. The functions  $O(r)$  and the extinction term  $\alpha(r)$  given by Eq. (5.4) are the same in the two equations. Once the two constants  $K_{\text{mol}}$ ,  $K_{\text{aer}}$  or, rather, their ratio  $K = K_{\text{mol}}/K_{\text{aer}}$  have been determined, Eqs. (5.7) and (5.8) can be divided, directly yielding the lidar backscatter ratio

$$\Re(r) = \frac{\beta_{\text{aer}}(r)}{\beta_{\text{mol}}(r)} = \frac{K P_{\text{aer}}(r)}{P_{\text{mol}}(r)}. \quad (5.9)$$

High spectral resolution lidars utilize optical filters to distinguish between photons scattered from molecules and those scattered by aerosol or cloud particles. Very narrow bandwidth filters are required ( $\sim 1$  GHz). In addition, the transmitting laser frequency must be locked to the filter center frequency, and the linewidth must be smaller than the filter width ( $\sim 100$  MHz). These requirements make HSRLs more difficult to implement than Raman lidars. However, a HSRL provides much larger molecular signals and can utilize very narrow bandwidths to block solar noise.

## 5.3 HSRL Implementations

### 5.3.1 Scanning Fabry–Perot Interferometer

The combined particulate and molecular spectrum can be observed using any frequency-stabilized laser and a scanning Fabry–Perot interferometer. The technique was first proposed and demonstrated by Fiocco et al. [4] using a line-narrowed cw-argon-ion laser and a scanning Fabry–Perot interferometer.

The broadband molecular component of the measured spectrum can be fitted to predictions of a model molecular spectrum. The backscatter ratio can then be determined from the atmospheric density at the measurement altitude and the ratio of the areas under aerosol and molecular scattering curves. Because the filter bandwidth is typically much narrower than the molecular spectrum, the filter rejects most of

the molecular light. This makes the system efficiency low and the measurement time long. Spectral components are measured sequentially, allowing temporal variations of the atmospheric conditions to distort the spectrum. As a result, the spectral scanning approach is unattractive for most atmospheric measurements.

### 5.3.2 Fixed Fabry–Perot Interferometer

System efficiency can be improved with nonscanning Fabry–Perot systems [5, 6]. In this case the Fabry–Perot etalon is locked to the laser wavelength. Two detectors are employed as shown in Fig. 5.2. One measures the signal passing through the etalon and the other measures the reflected signal. Most of the particulate signal passes through the etalon with only a small fraction reflected. Meanwhile, the Doppler-broadened molecular signal is divided more equally between detectors. If the spectral transmission and reflection characteristics of the etalon are known, a model of the molecular spectrum can be used with an independently supplied atmospheric temperature profile to predict the transmission of the two channels for both particulate and molecular signals. The measured signals in the two channels can then be expressed as linear combinations of the photons scattered from particulate matter and from gas molecules. These equations can be inverted to separate the molecular and particulate component, as is shown in Subsection 5.4.3. For more details see Sroga et al. [7] and Grund et al. [6].

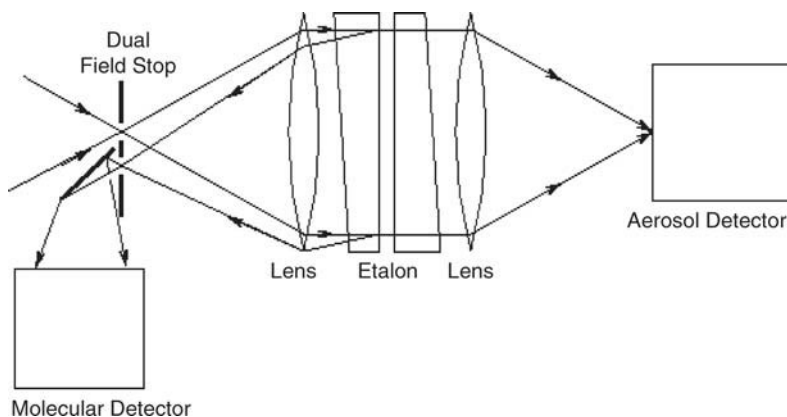


**Fig. 5.2.** An etalon-based HSRL. The etalon forms a narrow-band filter for light transmitted to the aerosol detector. Light reflected from the etalon is directed to the molecular detector. A pre-filter (not shown) is used to suppress skylight.

The nonscanning etalon approach has several advantages: (1) Errors due to temporal variations in the scattering media are suppressed because both signals are measured simultaneously. (2) System efficiency is improved because the filter bandwidths are larger, and both transmitted and reflected signals are detected. (3) The etalon can be tuned to any wavelength. For example, the system described by Shipley et al. [5] was tuned to operate at the wavelength of an iron Fraunhofer line in the solar spectrum where background solar light is reduced by approximately a factor of five. Or in the case of the system described by Grund et al. [6], the etalon allowed use of a nontunable copper-chloride laser. Also, the lidar may be operated at UV wavelengths, improving eye safety. The major disadvantage of the nonscanning approach lies in the sensitivity of high-resolution etalons to thermal and mechanical perturbations. In addition, at a given spectral resolution the product of the etalon diameter and the angular acceptance of an etalon is limited. Reducing the telescope field-of-view (FOV) can decrease the angular cone of light incident on the etalon. However, practical constraints determine the smallest FOV. The only alternative that will maintain the required spectral resolution as the telescope diameter is increased is to increase the diameter of the etalon plates. As a result, large telescopes require large, expensive etalons. For example, the systems described in Shipley et al. [5] and Grund et al. [6] (350-mm telescopes, 350- $\mu$ rad FOV), required 150-mm-diameter etalons.

### 5.3.3 Atomic and Molecular Absorption Filters

Atomic and molecular absorption filters offer an attractive alternative to Fabry–Perot-based systems. Researchers at Colorado State University pioneered this approach using barium vapor filters [8–10]. A dye laser transmitter tuned to a Ba atomic absorption line at a wavelength of 553.7 nm was used in conjunction with heated (700–800 °C) absorption cells containing Ba vapor. Light from the receiving telescope was directed through a beamsplitter with one part of the light sent directly to a detector while the rest of the light was directed through the Ba cell as shown in Fig. 5.3. The central peak of the molecular spectrum and all of the particulate scattering is absorbed, allowing only the spectral wings of the molecular scattering to pass through to a second detector.

The system proposed by Shimizu et al. [8] was designed to simultaneously provide atmospheric temperature measurements and measurements of backscatter coefficients and optical depths. A second

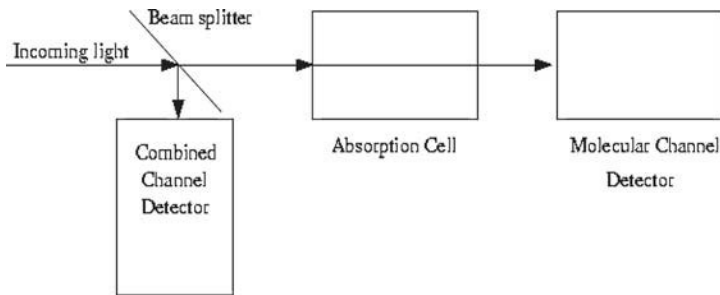


**Fig. 5.3.** HSRL detector configuration when using an atomic or molecular absorption filter. The absorption filter is evacuated and then filled with the absorbing gas. It is normally enclosed in a temperature-controlled housing to minimize sensitivity to environmental temperature changes. It is important to control the polarization of the incoming light to prevent calibration errors caused by the polarization sensitivity of the beam splitter.

beam splitter installed behind the first directed part of the light through a second Ba cell. The second cell was maintained at a higher temperature than the first cell in order to increase its spectral width. Because the spectral width of the Doppler-broadened molecular backscatter is proportional to the square root of the temperature, the ratio of the signals seen by the detectors behind the two Ba cells is a function of the atmospheric temperature. The change in signal ratio due to temperature is small, making this a difficult measurement. Additional information on temperature measurements using Ba absorption cells can be found in papers by the Colorado State research group [8–10].

The atomic vapor version of the HSRL replaces the temperature-sensitive and mechanically sensitive Fabry–Perot with a robust and stable atomic absorption filter. It also avoids the acceptance angle limitations of the Fabry–Perot. The major disadvantages of the Ba filter are high operating temperatures and lack of a convenient laser source emitting at the barium absorption wavelength. The Colorado State University group used a Nd:YAG-pumped dye laser as a transmitter in conjunction with the Ba vapor filter.

A much simpler implementation of HSRL is achieved when the Ba cell is replaced with a molecular iodine cell as described by Piironen et al. [11, 12]. (Related information on the use of iodine absorption cells in wind tunnel Doppler velocimetry is found in Forkey [13].) The iodine absorption cell shares the robust spectral stability and wide acceptance angle of the Ba cell while allowing operation at much lower temperatures ( $\sim 25$  to  $\sim 100^\circ\text{C}$ ). In addition, it has several suitable absorption



lines within the thermal tuning range of the 532-nm frequency-doubled Nd:YAG laser. (A catalog of iodine absorption lines is provided by Gerstenkorn and Luc [14].) CW-diode-pumped seed lasers are available to injection-lock the laser and generate the necessary narrow spectral line width for the transmitter.

A pair of iodine absorption cells held at different temperatures can also be used to make atmospheric temperature measurements. This work is described in Hair et al. [15] and a PhD thesis by Hair [16].

## 5.4 HSRL Designed for Remote Operation

The latest University of Wisconsin HSRL employs an iodine absorption cell to separate the molecular signal. It is designed for long-term unattended operation in the Arctic where it will be controlled remotely and operate as an Internet appliance. Use of a high-repetition-rate laser and expansion of the transmitted beam through a 400-mm telescope reduces the transmitted energy density to eye-safe levels. It is possible to look directly into the output beam without hazard. Using the same telescope for the transmitter and receiver makes it easy to maintain stable alignment of the transmitter and receiver although the angular FOV is only  $45\text{ }\mu\text{rad}$ . The small FOV and the 4-kHz repetition rate also limit the near-field signal strength, making it possible to record continuous profiles that start at an altitude of  $\sim 50\text{ m}$  and extend to  $30\text{ km}$  using photon counting detectors. The small FOV also suppresses multiple-scattering contributions. Table 5.1 lists the technical data, Fig. 5.4 presents a sketch of the University of Wisconsin unattended HSRL system.

**Table 5.1.** UW arctic HSRL technical data

|                        |                           |                         |                       |
|------------------------|---------------------------|-------------------------|-----------------------|
| Average transmit power | 600 mW                    | Optical detectors       | Geiger-mode APDs, PMT |
| Pulse repetition rate  | 4 kHz                     | APD quantum efficiency  | $\sim 60\%$           |
| Wavelength             | 532 nm                    | PMT quantum efficiency  | $\sim 5\%$            |
| Solar noise bandwidth  | 8 GHz                     | Data acquisition        | Photon counting       |
| Angular field-of-view  | $45\text{ }\mu\text{rad}$ | Range resolution        | 7.5 m                 |
| Telescope diameter     | 400 mm                    | Maximum time resolution | 0.5 s                 |

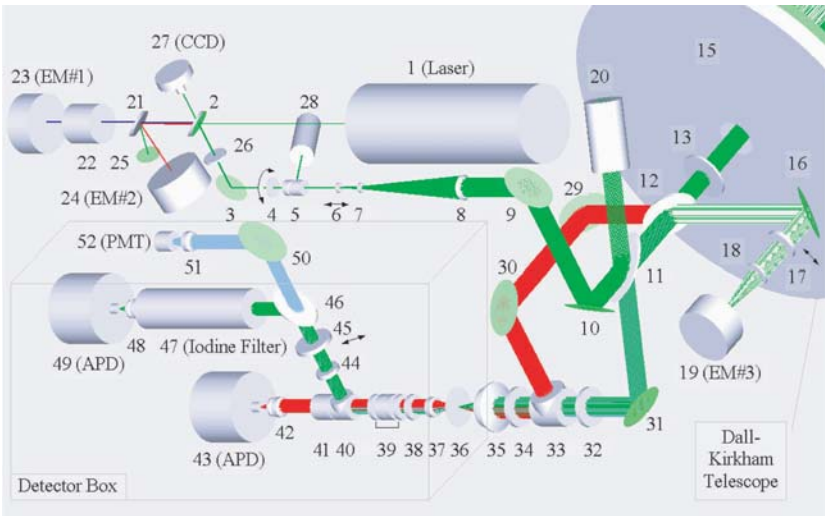


**Fig. 5.4.** Schematic showing the University of Wisconsin Arctic HSRL system. Component descriptions are provided in Table 5.2. The cross-polarized beam path is shown in red.

**Table 5.2.** HSRL component descriptions

|                                                |                                     |
|------------------------------------------------|-------------------------------------|
| 1 Laser                                        | 27 CCD camera                       |
| 2 Wedged beamsplitter                          | 28 Light trap                       |
| 3 Mirror                                       | 29, 30, 31 Mirrors                  |
| 4 Half-wave plate                              | 32, 34 Half-wave plates             |
| 5 Glan linear polarizer                        | 33 Polarizing beamsplitter          |
| 6, 7, 8 Beam expander                          | 35 Gradium + meniscus lens          |
| 9, 10 Mirrors                                  | 36 50- $\mu$ m field stop           |
| 11 Thin-film polarizer                         | 37 Plano-convex lens                |
| 12 8% mirror                                   | 38 Interference filter (.35 nm)     |
| 13 Quarter-wave plate                          | 39 8-GHz, air-spaced etalon         |
| 14 Telescope secondary (not shown)             | 40, 41 Polarizing beamsplitter      |
| 15 Telescope primary                           | 42 Gradium+meniscus lens            |
| 16 Black glass flat                            | 43 APD detector (dia = 170 $\mu$ m) |
| 17 Removable ND filter, OD = 3                 | 44 Half-wave plate                  |
| 18 Plano-convex lens                           | 45 Insertable OD = 3 filter         |
| 19 Transmitted-energy monitor                  | 46 Beamsplitter                     |
| 20 Light trap                                  | 47 272-mm long I <sub>2</sub> cell  |
| 21 Wedge                                       | 48 Gradium + meniscus lens          |
| 22 Calibration iodine cell                     | 49 APD detector                     |
| 23, 24 I <sub>2</sub> -locking energy monitors | 50 Mirror                           |
| 25 Mirror                                      | 51 Plano-convex lens                |
| 26 Mirror, R = 1/2%                            | 52 PMT detector                     |

### 5.4.1 Transmitter

The laser transmitter (1) is a diode-pumped, intracavity, frequency-doubled Nd-YAG laser. Narrow-band, single-frequency operation is provided by injection seeding with a single-frequency, cw-diode-pumped diode laser. The main laser cavity is maintained in resonance with the seed laser by adjusting the cavity length to minimize the time between the Q-switch trigger and the emission of the laser pulse. The emission wavelength is tuned via temperature control of the seed laser crystal and is locked to line #1109 of the iodine absorption spectra (line numbers from [14]). Locking is accomplished by minimizing the transmission through a 2-cm-long iodine absorption cell (22). Energy monitors (23) and (24) are used to measure the transmission. A half-wave plate (4) mounted in a computer-controlled rotation stage and a Glan-Thompson polarizer (5) allow computer control of the emitted power without changing the operating characteristics of the laser; this power control is used during system calibration.

A small beam expansion telescope (6–8) enlarges the  $\sim 1$ -mm laser beam to overfill a 20-mm aperture. Overfilling wastes some of the laser power but serves to flatten the energy distribution in the beam. This allows more energy to be transmitted without exceeding eye-safety limits at any point within the output aperture. The linearly polarized transmit beam is converted to circular polarization by a quarter-wave plate (13) and then expanded by a  $20\times$  afocal telescope (15) to a final diameter of 400 mm.

CCD camera (27) images interference fringes formed by reflections from the mirror (25) and a partially reflective mirror (26). The motion of these fringes is used to determine the laser frequency while tuning the laser during calibration. An absolute calibration of the fringe information is derived from known positions of the iodine absorption lines.

### 5.4.2 Receiver

The receiver and transmitter use the same afocal telescope (14,15). This greatly simplifies the task of maintaining alignment between the transmitted beam and the receiver field of view and permits the lidar to operate with a  $45\text{-}\mu\text{rad}$  FOV. The telescope directs the received photons through the quarter-wave plate (13). Received photons that have maintained their polarization (except for the reversal which occurs on changing their direction of propagation) are converted into linear polarization with its axis of polarization perpendicular to that of the transmitted photons. A portion of these photons and part of the photons whose polarization has

been changed by the scattering media are separated by a beamsplitter comprised of a glass plate (12) that is antireflection coated on one surface and uncoated on the other. This beamsplitter directs 12% of the cross-polarized photons to the input of a polarizing cube (33) where the beam is combined with the rest of the signal for transmission through the field stop and the background noise filter. This signal is used to measure depolarization of the return signal. Meanwhile, the photons that did not have their polarization changed by scattering and have managed to pass through the cross-polarization pick-off (12) encounter the thin-film polarizer that forms the transmit/receive switch (11) and are directed to the other entrance face of the polarizing cube (33). This element combines parallel and cross components of polarization into a single beam and maintains their orthogonal linear polarizations. The combined beam is then focused by lens (35) onto the 50- $\mu\text{m}$ -diameter pinhole (36) that defines the receiver FOV. After collimation by lens (37), both beams pass through a skylight background filter comprised of a 0.35-nm-bandpass interference filter (38) followed by an 8-GHz-bandpass pressure-tuned etalon. Air pressure within the etalon chamber is computer-controlled via a stepper-motor-driven stainless-steel bellows. At this point polarization cube (40) separates the parallel-polarization signal from the cross-polarized component and passes the cross component on to a second polarizer (41) that removes any residual parallel polarization. The cross-polarized signal is then detected by a Geiger-mode avalanche photodiode (APD) photon-counting module (43). The APD is used in this channel because it offers higher quantum efficiency ( $\sim 60\%$ ) than is available in a PMT. A neutral density filter ( $\text{OD} = 3$ ) (44) is inserted into the optical path during calibration. This filter, plus a power reduction achieved via a rotation of the half-wave plate (4), reduces the number of photons scattered from optical surfaces in the lidar so that they can be counted without large pile-up corrections. This allows the scattered light pulse to be used in the calibration of the spectral bandpass of the lidar. The polarization sensitivity of the beamsplitter (46) along with the rotation of the half-wave plate (44) is used to balance signal strengths at the combined (52) and at the molecular (49) detectors. The beamsplitter sends part of the signal to a photon-counting PMT detector (52), which detects a signal containing both particulate and molecular scattered photons. A PMT is used here because it can accommodate high photon counting rates that are generated by dense, low-altitude clouds. The other component of the parallel-polarized light leaving the beamsplitter (46) is directed through a 272-mm iodine absorption cell (47) and focused

onto a second Geiger-mode APD photon-counting module (49). A high-QE APD detector can be used in this channel because the iodine filter blocks the intense particulate scattering signal from clouds. The entire system except for the telescope is housed in a temperature-controlled enclosure to minimize both thermally induced alignment changes and changes in the spectral characteristics of the iodine absorption cells. In addition, the iodine cells are operated at 30°C: this is  $\sim 2$  K above the temperature at which all of the iodine in the cell evaporates. The temperature sensitivity of the cell is reduced because the iodine vapor density does not change in response to condensation and evaporation of iodine.

### 5.4.3 Calibration

The signals  $S_c$  and  $S_m$  detected in the combined channel and in the molecular channel, respectively, can be described as a linear combination of the number  $N_a$  of aerosol photons and the number  $N_m$  of molecular photons incident on the lidar receiver:

$$S_c = N_a + C_{mc}N_m, \quad (5.10)$$

$$S_m = C_{am}N_a + C_{mm}N_m. \quad (5.11)$$

The equations have been normalized relative to the response of the combined channel to aerosol photons. This normalization is possible because an absolute calibration is generated by comparing the observed molecular lidar return to the lidar return computed from Rayleigh scattering theory.  $C_{mc}$  describes the relative contribution of molecular photons to the combined channel.  $C_{mc}$  may be less than unity if the pre-filter used to block sunlight is sufficiently narrow to affect the transmission of the Doppler-broadened molecular scattering.  $C_{am}$  describes the response of the molecular channel to aerosol photons and accounts for the on-line leakage of the absorption filter.  $C_{mm}$  describes the transmission of molecular photons through the absorption filter. Equations (5.10) and (5.11) can be inverted to compute the relative number of aerosol and molecular photons incident on the system:

$$N_a = \frac{C_{mm}S_c - C_{mc}S_m}{C_m - C_{am}C_{mc}}, \quad (5.12)$$

$$N_m = \frac{S_m - C_{am}S_c}{C_{mm} - C_{am}C_{mc}}. \quad (5.13)$$

The coefficients  $C_{am}$ ,  $C_{mm}$ , and  $C_{mc}$  are determined by directing a sample of the transmitted beam into the receiver while scanning the laser frequency. The spectral transmissions (see Fig. 5.5) for the molecular and combined channel are then convolved with the Doppler-broadened molecular spectrum, which is computed from theory and independently supplied temperature data.  $C_{mm}$  and  $C_{mc}$  are the values of the molecular and combined convolution functions at the operating frequency of the laser. Because  $C_{mm}$  and  $C_{mc}$  are temperature and pressure dependent, they must be computed as functions of altitude.  $C_{am}$  is taken directly from the measured leakage of the absorption filter at the operating frequency of the laser.

In the lower atmosphere, Brillouin scattering causes the Doppler-broadened molecular line shape to vary slightly from the Gaussian profile predicted for a Maxwellian velocity distribution. A line-shape model that

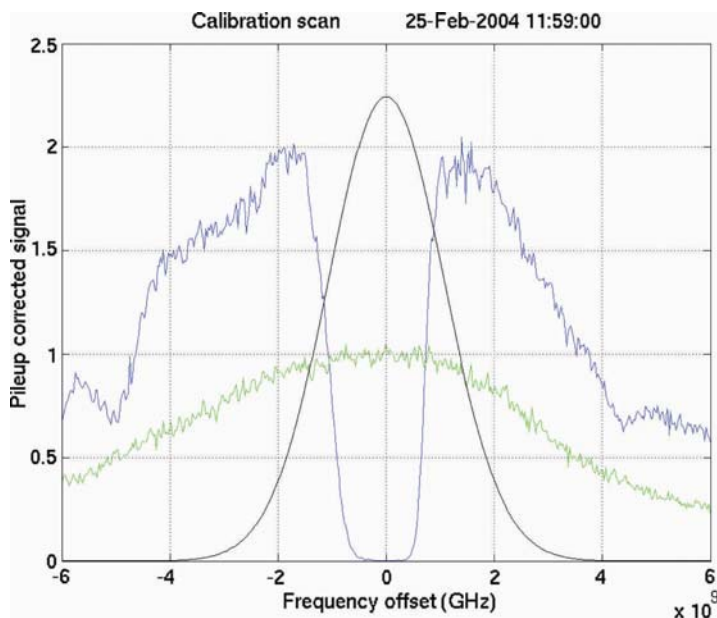


**Fig. 5.5.** Calibration scan showing the transmission of the molecular (blue) and combined (green) channels as a function of frequency. The Doppler broadened molecular spectrum for 300 K is also shown (black). Line 1109 of the iodine absorption spectrum (central notch) rejects most of the aerosol scattering and the central portion of the molecular scattering while passing the wings of the molecular line. The spectral transmission of the combined channel is determined by the pre-filter etalon.

includes Brillouin scattering is described by Tenti et al. [17] and a Fortran computer implementation can be found in Forkey [13].

## 5.5 Data Examples

Figures 5.6–5.11 present data acquired with the University of Wisconsin HSRL. Figure 5.6 shows an attenuated backscatter-coefficient image acquired on 14 January 2004. The data represent the output of the combined channel corrected for the squared distance, for laser pulse energy, and for the laser-beam FOV overlap function. They are not corrected for attenuation but normalized using the molecular signal to show the correct backscatter coefficient at an altitude of 75 m. A well-calibrated conventional lidar would produce an identical image. Note how the cirrus cloud at 7 km is shadowed by the 3.4-km water cloud that appears at 6:10 UTC. Also note the strong lidar return seen below the clouds as a result of the combined effect of aerosol and molecular scattering. Data gaps at 8 and 12 UTC occur during system calibrations.

Using the same data set, Fig. 5.7 presents the backscatter coefficient derived from both the molecular and combined-channel data. This image is calibrated absolutely, molecular scattering has been removed. Areas with insufficient signal for the HSRL inversion are indicated by black shadows; everywhere else, attenuation has been removed. Note the effect of attenuation correction on the appearance of the 7 km cirrus cloud. The lower cloud no longer affects the scattering coefficient measured above.

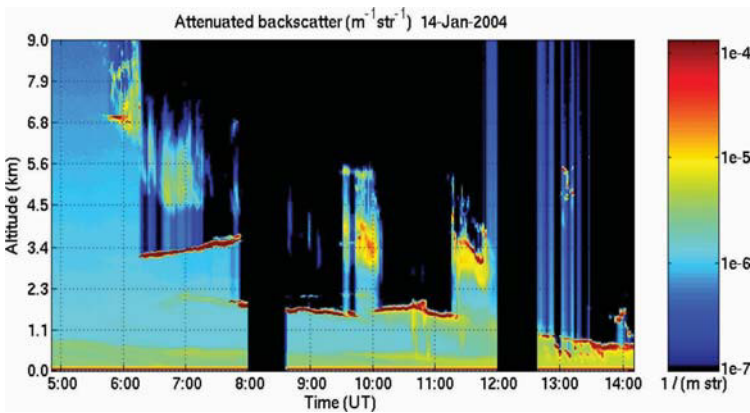


**Fig. 5.6.** Attenuated backscatter image recorded with the HSRL on 14 January 2004.

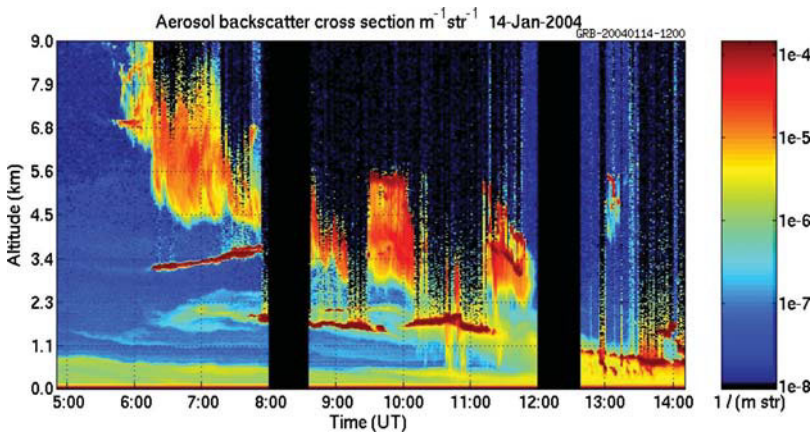


**Fig. 5.7.** Aerosol backscatter coefficient recorded on 14 January 2004.

As long as sufficient signal is available the measurements are unchanged. Also note how removal of the molecular scattering increases contrast in the aerosol structure below the clouds (i.e., at 2 km and 7 UTC).

The UW Arctic HSRL transmits circularly polarized light. The receiver separates the returned signal into two left and right-handed components. Light scattered from spherical particles is not depolarized and is returned to the combined and molecular channels. A portion of

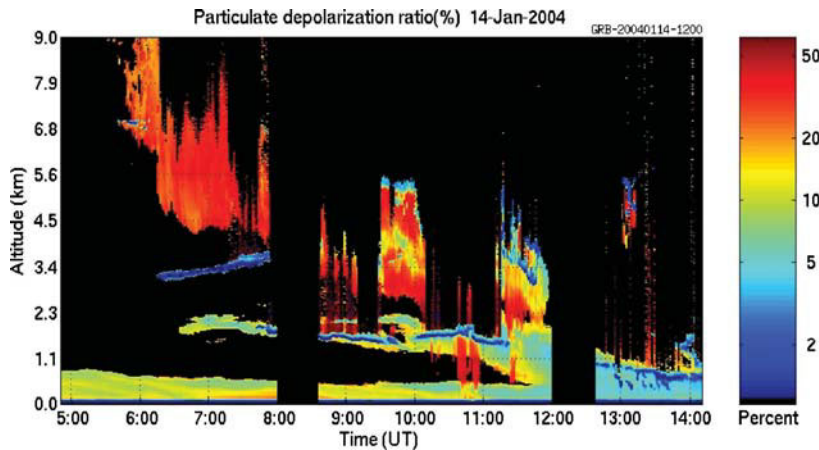


**Fig. 5.8.** Circular depolarization ratio recorded on 14 January 2004. Note the logarithmic scale.



the light scattered by nonspherical particles is depolarized. The cross-polarized channel measures this signal. Figure 5.8 shows the circular depolarization ratio computed from these signals. As expected, light backscattered from water clouds maintains the transmitted polarization and exhibits very low depolarization ( $<1\%$ ) while ice clouds show high depolarization ( $\sim 40\%$ ). Aerosol depolarization often varies between 0 and 15% depending on the type of aerosol; water hazes exhibit very low depolarization while irregularly-shaped dust particles show larger values.

Figure 5.9 shows the aerosol backscatter ratio measured in the relatively clear period before 5:30 UTC. These values were computed from the calibration scan and a radiosonde temperature profile. Near 9 km the scattering-ratio profile shows a minimum value of  $(-0.024)$ . This layer is likely to be nearly aerosol-free due to scavenging by the cirrus cloud above. The negative value gives an indication of the error present. It is due to a combination of lidar characterization errors and errors in the temperature profile. The temperature profile was obtained from a

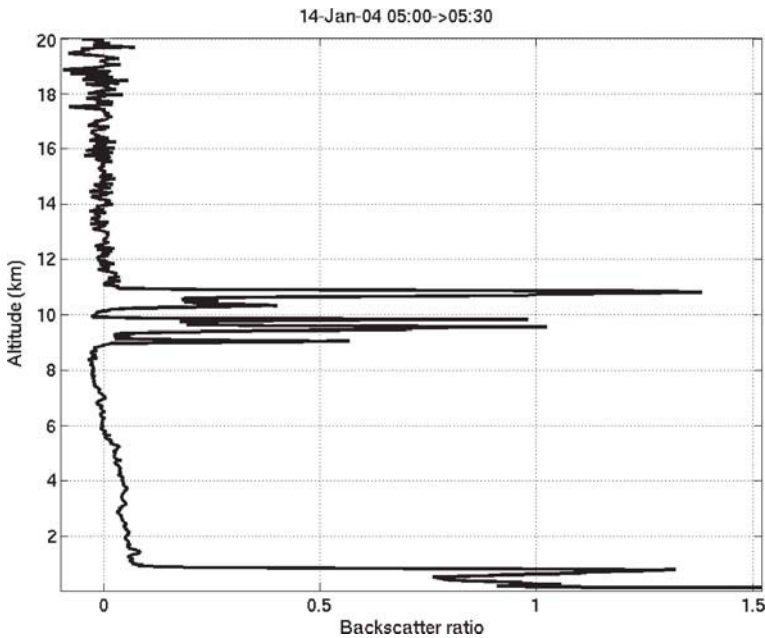


**Fig. 5.9.** Scattering ratio profile measured between 5:00 and 5:30 UTC on 14 January 2004.

radiosonde launched about 5 hours earlier at a weather station approximately 150 km from the lidar. The HSRL technique makes it possible to compute rigorous error estimates [12]. However, this is a new lidar and system characterization is still in progress so this has not been completed.

Figure 5.10 shows backscatter coefficient profiles measured between 6:40 and 6:41 UTC. The basis of the HSRL optical depth measurement is easily seen in comparing the attenuated molecular profile to the Rayleigh backscatter profile computed from radiosonde data. The wide dynamic range of the lidar is also evident. This figure shows measurements ranging over five decades. When the time averaging is increased to a few minutes, the instrument can provide measurements over a range of more than six decades. Careful data processing is required to maintain accuracy over this wide dynamic range. Background sky noise and dark counts must be subtracted. Pileup correction must be applied to properly represent signals at high count rates. Detector afterpulse corrections

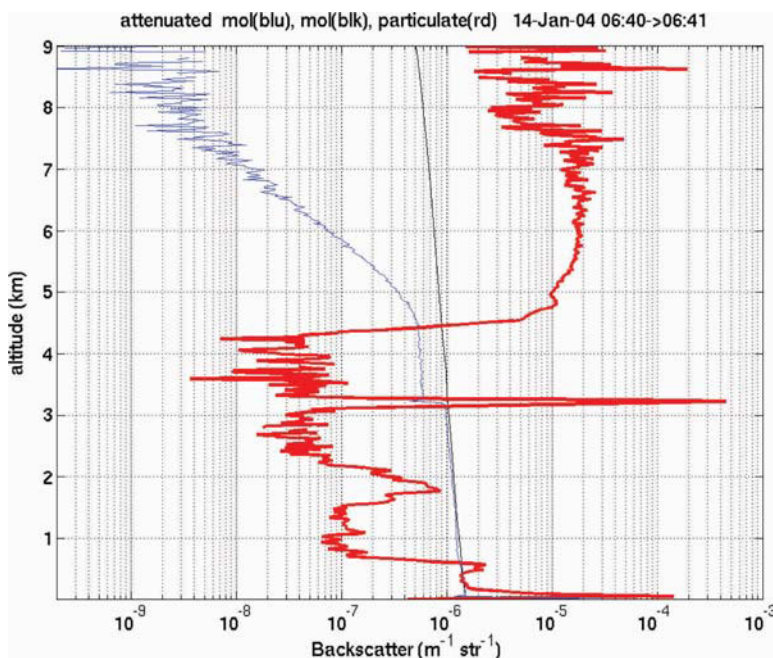


**Fig. 5.10.** The one-minute average aerosol backscatter-coefficient profile (red) and the attenuated molecular return (blue) observed between 6:40 and 6:41 UTC. The Rayleigh return computed from the 00 UTC Green Bay, Wisconsin, radiosonde is shown in black.

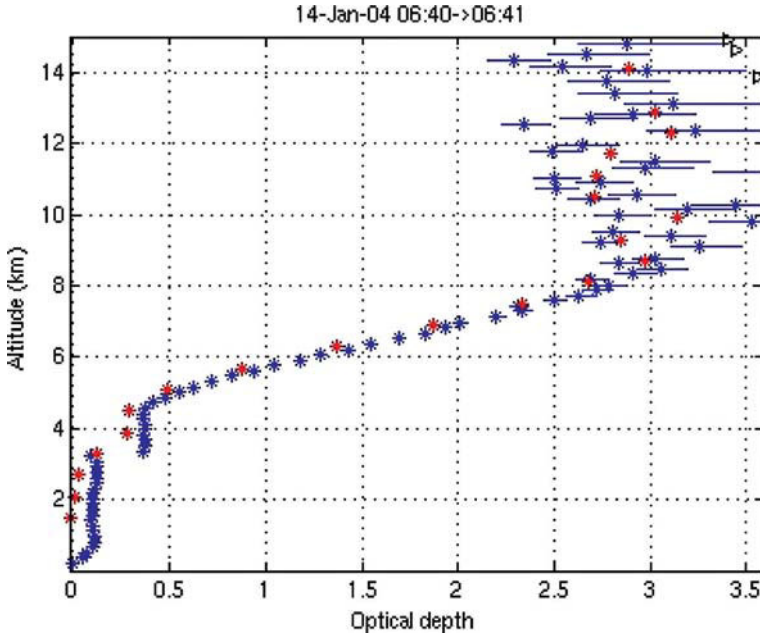


**Fig. 5.11.** Optical depth profiles derived for a one-minute time average and altitude averages of 150 m (blue) and 600 m (red).

must be carried out to remove spurious counts generated by previously detected photons [12].

Figure 5.11 shows the optical depth as a function of altitude derived for a 1-minute average between 6:40 and 6:41 UTC. These data were computed using 150 m (blue) and 600 m (red) vertical averages. Uncertainty estimates based on photon counting errors are shown. In this case, photon statistics dominates the noise. Longer time averages reduce the statistical error. However, other factors, such as detector after-pulsing, limit the maximum optical depth that can be measured to  $\sim 4$ .

## 5.6 Future Prospects

HSRL, if built along the lines followed in the design of the University of Wisconsin Arctic High-Spectral-Resolution Lidar, offers a unique combination of advantages: (1) robust calibration and no unstable inversions or a-priori assumptions required to derive scattering coefficients and optical depths; (2) eye-safe operation that allows direct viewing of

the transmitted beam while operating at the wavelengths most important for human vision and most important for the transfer of solar radiation in the atmosphere; (3) photon counting detectors allowing simple robust calibration; (4) large dynamic range allowing the system to operate without regard to atmospheric conditions—no operator is required to change parameters in response to varying atmospheric conditions; (5) narrow angular field-of-view that limits multiple scattering in water clouds to negligible levels and reduces background sky noise; (6) automatic data processing possible because the signals contain sufficient information to derive products without ambiguity; (7) profiles normalized to atmospheric molecular scattering and independent of window transmission—the lidar continues to make accurate measurements even though the external window transmission is degraded by water, snow, or dirt.

Despite these advantages, few HSRL systems have been constructed; technical complexity has limited deployment. A single-mode laser with a spectral linewidth below 100 MHz is required. Separation of the aerosol and molecular return requires a filter with a bandwidth of approximate 1 GHz. The laser and the filter must be frequency-locked to within approximately 100 MHz. High repetition rate systems also require an etalon pre-filter to limit skylight and very narrow field-of-view receivers with stringent requirements to maintain filter tuning and transmitter/receiver alignments.

Advances in technology have greatly reduced the challenges associated with the HSRL technique. High repetition rate, injection-seeded, diode-pumped Nd:YAG lasers are available as standard commercial products. These lasers consume a few hundred watts of power and operate for thousands of hours without maintenance. They replace flashlamp-pumped lasers that used kilowatts of power and required maintenance at  $\sim 100$ -hour intervals. Simple, robust and stable, iodine absorption filters replace the environmentally sensitive Fabry–Perot etalon used to separate the molecular and aerosol returns. Photomultiplier tubes are replaced by Geiger-mode avalanche-photodiodes as high-quantum-efficiency photon-counting detectors in simple-to-use packages.

In the near future, fiber amplifiers will boost the power of miniature narrow-band lasers to the levels required by HSRL systems. These will further reduce the power consumption, volume, and cost of the transmitter. They will operate for tens of thousands of hours without maintenance and will eliminate the requirement for water cooling. It also appears

likely that iodine absorption filters will be constructed with hollow-core photonic-crystal fibers and that fiber components will replace many of the traditional optical elements in the receiver.

Until now, HSRLs have been in use at a limited number of research laboratories. The intrinsic advantages of the HSRL technique, coupled with advancing technology, are likely to greatly expand its deployment and to make high spectral resolution lidar a routine observational instrument in service at numerous automated weather stations and airports around the world.

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## Visibility and Cloud Lidar

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### 6.1 Introduction

Visibility, or visual range, is a property of the atmosphere that has direct significance not just for our pleasure and well-being. Visibility is of decisive importance for all kinds of traffic operations. An uncounted number of victims have been injured or killed because of visibility or, rather, the lack of it. In road traffic a cautious driver can go slowly and stop altogether if, in a snowstorm or in heavy rain or fog, visibility gets worse and worse. This possibility is restricted in boat operations and absent in air traffic.

Unlike *in situ* devices which determine visibility in one point, or fixed installations for the determination of optical transmission between two locations on the ground, lidar allows one to make observations of atmospheric conditions over an extended optical path from one location, fixed or mobile, in any, not just in horizontal direction.

Lidar allows quantitative determination of visual range as a function of distance and is thus ideally suited to measure visibility at airports both over the different parts of runways and in the air space above where aircraft safety depends particularly critically on the pilots' unimpeded view and orientation.

## 6.2 The Notion of Visual Range

Visual range is in this context defined as a local variable of the atmosphere that is directly related to its turbidity: the higher turbidity, the shorter the visual range.

Visibility is physically limited by two effects: the inability of light from a distant object to reach the eye of the observer due to atmospheric absorption, and the increase of background light from atmospheric scattering between the object and the observer [1]. In other words: an object can no longer be seen by the observer if its apparent brightness gets close to the brightness of the background—exactly how close is a matter of definition as will be shown below.

The magnitude of the former effect is described by the atmospheric absorption coefficient  $\alpha(x, \lambda)$  which may depend on range  $x$  and which varies with wavelength  $\lambda$ . The magnitude of the latter is determined by the atmospheric backscatter coefficient  $\beta(x, \lambda)$ . Visibility also depends on the position of the sun, on the color of the object, and on other parameters.  $\alpha$  and  $\beta$  are related to each other. For molecular scattering they are strictly proportional, the proportionality factor being  $\alpha/\beta = (8\pi/3)$  sr. For particle (cloud and aerosol) scattering they are still proportional, but the factor varies by an order of magnitude with aerosol material, particle size distribution, moisture, and other aerosol properties.

According to Koschmieder's theory [2], visual range  $V$  is determined only by the contrast threshold  $K$  an observer needs to distinguish an object from its background, and by the extinction coefficient  $\alpha$ .

Variations in the relation between  $\alpha$  and  $\beta$  and all other effects mentioned are neglected in the simple definition

$$V(x) = \frac{1}{\alpha(x)} \ln \frac{1}{K}. \quad (6.1)$$

The *average* visual range between two points at distance  $x_1$  and  $x_2$  is then given by the obvious relation

$$\bar{V} = \frac{x_2 - x_1}{\int_{x_1}^{x_2} \alpha(\xi) d\xi} \ln \frac{1}{K}. \quad (6.2)$$

Clearly, Eq. (6.1) defines a local and Eq. (6.2) an averaged atmospheric property.

### 6.2.1 Normal Visual Range

The convention is that  $\alpha$  is taken at  $\lambda = 550$  nm. At this wavelength visual sensitivity is best. Koschmieder [2] sets  $K = 0.02$ , the contrast threshold for a normal-sighted, experienced observer. From Eq. (6.1) we thus obtain the normal visual range, or normal optical range

$$\text{NOR} = \frac{1}{\alpha} \ln \frac{1}{0.02} = \frac{3.912}{\alpha}. \quad (6.3)$$

### 6.2.2 Meteorological Optical Range

For practical purposes a more conservative contrast threshold  $K = 0.05$  is assumed, taking into consideration psychological and stress effects to which an observer such as an aircraft pilot may be exposed, leading to the shorter so-called meteorological visual range (or meteorological optical range)

$$\text{MOR} = \frac{1}{\alpha} \ln \frac{1}{0.05} = \frac{3}{\alpha}. \quad (6.4)$$

The definition of *average* NOR and average MOR between two points is fully analogous to the definition of Eq. (6.2).

### 6.2.3 Vertical Optical Range

Horizontal stratification, or a marked variation of the extinction coefficient with height, is a frequent phenomenon. An observer looking up from the ground can see an object up to a height VOR defined, in analogy to the previous definitions, by the relation

$$\int_0^{\text{VOR}} \alpha(z) dz = 3. \quad (6.5)$$

Vertical optical range VOR is, in other words, the height above ground up to which the extinction coefficient  $\alpha$  must be integrated to yield the value 3; from this height one-twentieth of the light reaches an observer on the ground. Or, more important, 1/20 of the light generated at the ground reaches an observer at height VOR (which could be a balloon or aircraft pilot).



### 6.2.4 Slant Optical Range

Suppose an observer is at a height  $h$  above some horizontal plane, which can be a runway or, more generally, the ground. Slant optical range (SOR) is the maximum *horizontal* distance from the point exactly below him on the ground to another point on the ground he can see from his position at height  $h$ . With Pythagoras' theorem, we have

$$\text{SOR} = h \left[ \left( \frac{3}{\int_0^h \alpha(z) dz} \right)^2 - 1 \right]^{1/2}. \quad (6.6)$$

Slant optical range is thus the projection of a slant line of vision onto the horizontal plane. Clearly, SOR depends on height  $h$ . In most cases SOR decreases as  $h$  increases. When  $h$  reaches and exceeds VOR, the radicand in Eq. (6.6) gets negative, and SOR vanishes.

### 6.2.5 Runway Visual Range, Slant Visual Range

For the sake of completeness two more quantities are mentioned here, runway visual range (RVR) and slant visual range (SVR) (as opposed to SOR). These quantities are defined by the International Civil Aviation Organization ICAO [3, 4]. RVR is the maximum distance out to which a pilot, from a position 5 m above the runway, can recognize either the runway center line or the lights along the runway, defined again by the projection of the actual optical paths as in Eq. (6.6) onto the runway center line. In daylight, unless the runway lights are very bright, RVR=MOR.

In a similar way a quantity called slant visual range is defined for positions  $h \gg 5$  m. Again, if there are no bright landing fires, then SVR=SOR in daylight conditions.

## 6.3 Visibility Measurements with Lidar

Because of its great importance in air traffic, visibility was one of the first atmospheric quantities to which the lidar technique was applied [5]. As any lidar, visibility lidar also suffered from the problem that two atmospheric quantities, the absorption coefficient  $\alpha$  and the backscatter coefficient  $\beta$ , were to be determined from one measured quantity, the lidar signal.

Attempts to work at different measurement angles [6], use several wavelengths [7], and utilize reflections from hard targets [8] did not prove applicable. More sophisticated apparatus such as Raman and high spectral resolution lidar (HSRL) systems were not used because of their complexity in design and operation. The problem was finally solved by using essentially the same procedure as that described in Chapter 4, with the simplification that no separate knowledge of the aerosol and molecule contributions to  $\alpha$  and  $\beta$  is necessary, only the total values of the respective quantities, or even of total  $\alpha$  alone.

The method starts from the familiar lidar equation

$$P(x, \lambda) = \frac{c \Delta t}{2} P_0 \frac{A \eta O(x)}{x^2} \beta(x, \lambda) \tau^2(x, \lambda) \quad (6.7)$$

with  $P(x, \lambda)$  representing the signal power from distance  $x$  at wavelength  $\lambda$ ,  $P_0$  the average laser power transmitted during the pulse duration  $\Delta t$ ,  $A$  and  $\eta$  the receiver area and efficiency,  $O$  the laser-beam receiver-field-of-view overlap integral,  $\beta$  the backscatter coefficient, and

$$\tau(x, \lambda) = \exp \left[ - \int_0^x \alpha(\xi, \lambda) d\xi \right] \quad (6.8)$$

the one-way extinction between the lidar and the distance of interest,  $x$ . As simple visibility lidars work at one frequency only, we drop  $\lambda$  in the respective quantities and rewrite Eq. (6.7) to yield

$$P(x) = k \frac{O(x)}{x^2} \beta(x) \tau^2(x). \quad (6.9)$$

We define the quantity

$$S(x) \equiv \frac{P(x)x^2}{kO(x)} = \beta(x) \tau^2(x) \quad (6.10)$$

and solve the differential equation

$$\frac{\partial \ln(S(x))}{\partial x} = \frac{1}{\beta(x)} \frac{\partial \beta(x)}{\partial x} - 2\alpha(x) \quad (6.11)$$

to obtain, with the well-known approximations, the solution

$$\alpha(x) = \frac{S(x)}{\frac{S(x_m)}{\alpha(x_m)} + 2 \int_x^{x_m} S(\xi) d\xi} \quad (6.12)$$

in a procedure fully analogous to the one described in Chapter 4. It must be noted that in the present application the assumptions and approximations made in the solution of Eq. (6.11) hold sufficiently well. Equation (6.12) is thus a stable solution of the profile of the extinction coefficient  $\alpha(x)$ . To use it, however, the boundary values of  $S$  and  $\alpha$  at the remote end of the lidar range,  $S(x_m)$  and  $\alpha(x_m)$ , must be known. A data evaluation algorithm stable enough to also work in an automated regime is the following [9, 10]:

- (i) From the signal  $P(x)$  of the first measurement of a run, values are determined for the minimum and maximum range for which data evaluation appears meaningful.
- (ii) An initial value of  $\alpha(x_m)$  is chosen that is sufficiently large to lead to reasonably stable integration, but must be within the range of extinction coefficients that can be determined with the system. The lower limit of extinction coefficients is given by the signal-to-noise ratio, i.e., by the laser power and the sensitivity of the detector, the upper limit is determined by total extinction and thus, at fixed distance-bin width, by the number of bins.
- (iii) The integration is then carried out within the limits determined according to (i).
- (iv) Values of local visibilities are averaged, rejecting values beyond the limit predetermined in (ii).
- (v) The average is compared with the starting value. If the agreement is within 10%, it is considered the final value. If not, the average is taken for the next starting value, and steps (iii) to (v) are repeated. If this was not successful after 10 iterations, the procedure is stopped with an error message.
- (vi) If the iteration was successful, the resulting value is taken as a starting value for the next run, step (ii).

## 6.4 Aerosol Distributions

Impaired visibility is always caused by particles in the atmosphere. Particle distributions that frequently affect visibility are conveniently grouped in categories according to their origin, size, and effects. Particle size distributions span a wide range of radii  $r$  and are generally described by a modified gamma distribution

$$n(r) = ar^\varepsilon \exp(-br^\nu) \quad (6.13)$$

or by sums of such distributions each of which is then called a “mode” [11].  $n(r)dr$  is the number volume density of particles with radii between  $r$  and  $r + dr$ . The constants  $a$ ,  $b$ ,  $\varepsilon$ , and  $\gamma$  are all real and positive.  $\varepsilon$  and  $\gamma$  which describe the steepness of the rise and fall of the distribution are taken as integer and half-integer, respectively. The larger  $\varepsilon$  and  $\gamma$ , the steeper and narrower the mode. The radius of particles that are most abundant, or mode radius, is given by

$$r_c = \left( \frac{\varepsilon}{b\gamma} \right)^{\frac{1}{\gamma}}. \quad (6.14)$$

The total number density is obtained by integration over all particle radii:

$$N = a \int_0^{\infty} r^{\varepsilon} \exp(-br^{\gamma}) dr. \quad (6.15)$$

When carried out, the integration yields

$$N = a\gamma^{-1}b^{-(a+1)/\gamma}\Gamma((a+1)/\gamma). \quad (6.16)$$

The  $\Gamma$  function is related to the  $\Pi$  function which interpolates the factorials by the relation  $\Gamma(k) = \Pi(k-1)$ , with  $\Pi(0) = \Pi(1) = 1$ ,  $\Pi(2) = 2$ ,  $\Pi(3) = 6$ , etc.

In Table 6.1 nine common types of cloud, fog, and haze have been listed along with the parameters that describe particle size distributions if

**Table 6.1.** Parameters for the particle size distribution for several standard atmospheric conditions

| Atmospheric condition             | $a$                   | $\varepsilon$ | $b$    | $\gamma$ | $r_c$ ( $\mu\text{m}$ ) | $N$ ( $\text{m}^{-3}$ ) |
|-----------------------------------|-----------------------|---------------|--------|----------|-------------------------|-------------------------|
| Advection fog heavy               | 0.027                 | 3             | 0.3    | 1.0      | 10.0                    | $20 \cdot 10^6$         |
| Advection fog moderate            | 0.066                 | 3             | 0.375  | 1.0      | 8.0                     | $20 \cdot 10^6$         |
| Cumulus cloud C1/<br>Radfog heavy | 2.373                 | 6             | 1.5    | 1.0      | 4.0                     | $100 \cdot 10^6$        |
| Corona cloud C2                   | $1.085 \cdot 10^{-2}$ | 8             | 1/24   | 3.0      | 4.0                     | $100 \cdot 10^6$        |
| Haze H                            | $4.0 \cdot 10^5$      | 2             | 20.0   | 1.0      | 0.1                     | $100 \cdot 10^6$        |
| Haze L                            | $4.976 \cdot 10^6$    | 2             | 15.119 | 0.5      | 0.07                    | $100 \cdot 10^6$        |
| Haze M                            | $16/3 \cdot 10^5$     | 1             | 8.943  | 0.5      | 0.05                    | $100 \cdot 10^6$        |
| Cloud C3                          | 5.556                 | 8             | 1/3    | 3.0      | 2.0                     | $100 \cdot 10^6$        |
| Radfog moderate                   | 607.5                 | 6             | 3      | 1.0      | 2.0                     | $200 \cdot 10^6$        |

this distribution is assumed to be monomode. Four of these parameters, e.g.,  $a$ ,  $b$ ,  $\varepsilon$ , and  $\gamma$ , can be taken, independently from one another, from fits to measured distributions. The remaining two are then obtained from relations (6.14) and (6.16). In the table,  $\varepsilon$ ,  $\gamma$ ,  $r_c$ , and  $N$  were taken as primary parameters and  $a$  and  $b$  were calculated.

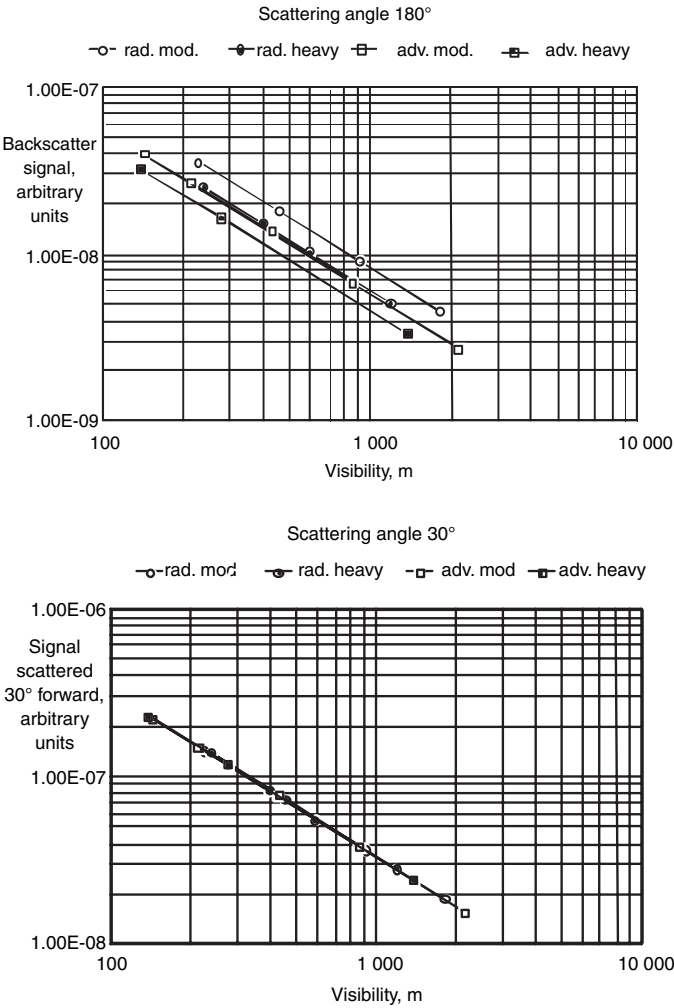


**Fig. 6.1.** Relative magnitude of scattered signal for four types of fog: radiative moderate, radiative heavy, advected moderate, and advected heavy. Whereas backscatter intensities (top) vary by a factor  $> 1.5$ , 30°-forward scattering is almost identical (bottom).

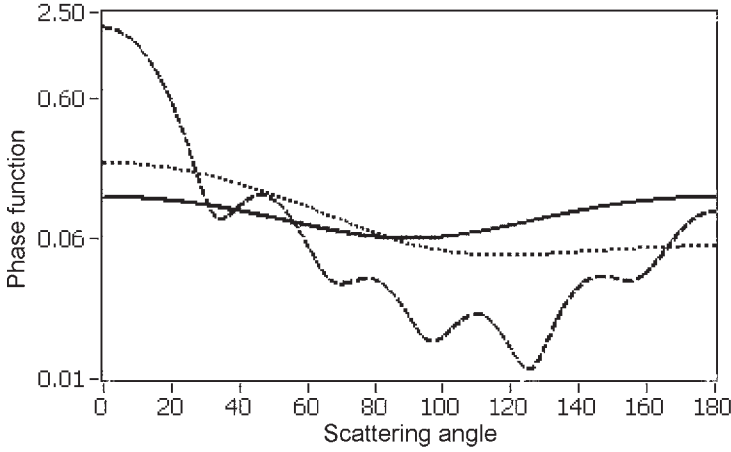


**Fig. 6.2.** Phase functions of spherical particles of different size. Solid line: 1 nm radius (Rayleigh scattering), dotted line: 100 nm, dashed line: 500 nm (Mie scattering). Scattering wavelength is 550 nm, index of refraction 1.55.

Much like the size distributions, the scattering phase functions—or scattering amplitudes—as a function of angle are also different for different weather conditions. For lidar, the scattering angle of relevance is  $180^\circ$ . Indeed, backscatter intensities vary by more than a factor of 1.5 for different fog conditions (Fig. 6.1, top). It is interesting to note that at a forward scattering angle of  $30^\circ$  these differences nearly vanish (Fig. 6.1, bottom). This is also seen in Fig. 6.2, which gives the scattering phase functions  $I$  for spherical particles of three different sizes. These functions are normalized such that the integral is unity:

$$\int_{\varphi=0}^{2\pi} \int_{\cos \vartheta=0}^1 I(\varphi, \vartheta) d \cos \vartheta d\varphi = 1. \quad (6.17)$$

## 6.5 Visibility and Multiple Scattering

In dense media, especially in clouds, the backscattered lidar signal may have undergone more scattering processes than just the near- $180^\circ$  backscatter process. Multiple scattering (Chapter 3) may strongly affect visibility measurements. The extent to which multiple scattering contributes to the lidar signal depends on the properties of the particles (size and volume number density, optical depth) and on the geometry of the lidar: the larger the volume from which light is detected, the

larger the multiple-scattering contribution. The fraction of multiply scattered light therefore increases with laser beam divergence, receiver field of view, and increasing distance between the lidar and the scattering volume.

Lidars are usually characterized by low beam divergence and a narrow receiver field of view. As large particles scatter light predominantly in the forward direction (cf. Fig. 6.2), the first scattering process occurs more often than for smaller particles in such a way that the scattered light is still within the lidar FOV so that it can directly be backscattered towards the receiver. Therefore large particles have the greatest share in multiple scattering.

Multiple scattering thus results in a reduction of the apparent extinction coefficient and a seemingly longer visual range. The effect is taken into account by appropriate correction terms for dense media.

The contribution of multiple scattering from dense media to the lidar return signal can be quite large. For illustration, Fig. 6.3 shows a simulated lidar return from a C1 cloud, 300 m thick, at a distance of 2000 m, and the relative contributions from single, double, and triple scattering events.

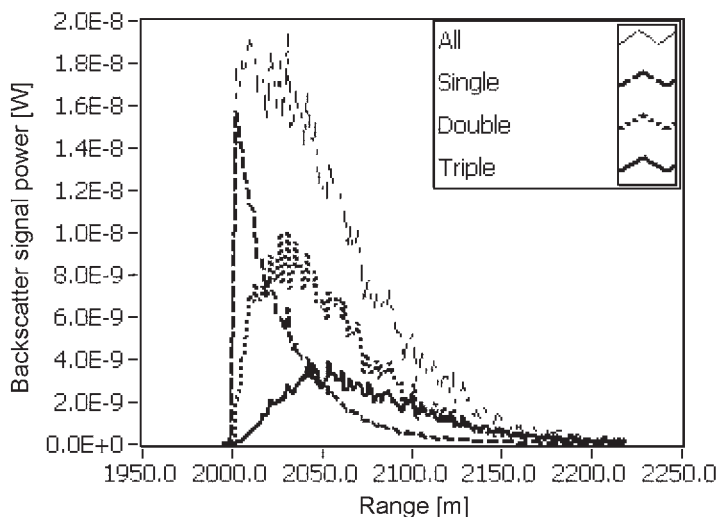


**Fig. 6.3.** Simulated lidar return signal from a C1 cloud of 300 m geometric depth at a distance of 2000 m. Dashed: single-scattering, dotted: double-scattering, solid: triple-scattering contribution to the total signal (light vertical bars).

## 6.6 Instruments

Visibility lidars are essentially backscatter lidars. Their sophistication is more in light weight, small volume, reliability, ruggedness, and ease of operation than in the ultimate in power, bandwidth, etc. Visibility lidar systems are commercially available from several manufacturers. Because they can also be used for other purposes such as cloud height detection and aerosol measurements, they often come under different names.

A family of systems particularly well suited for visibility measurements are the different types of Ceilometers provided by the Vaisala Company. Figure 6.4 shows such a system in operation at Oberpfaffenhofen, Germany.

The instrument transmits, at a wavelength around 900 nm, pulses of 30 W average power with a pulse repetition frequency of 6 kHz. The laser beam, expanded to 100 mm diameter, meets class-1M eye-safety criteria. One vertical aerosol profile (and thus one visibility profile) is produced

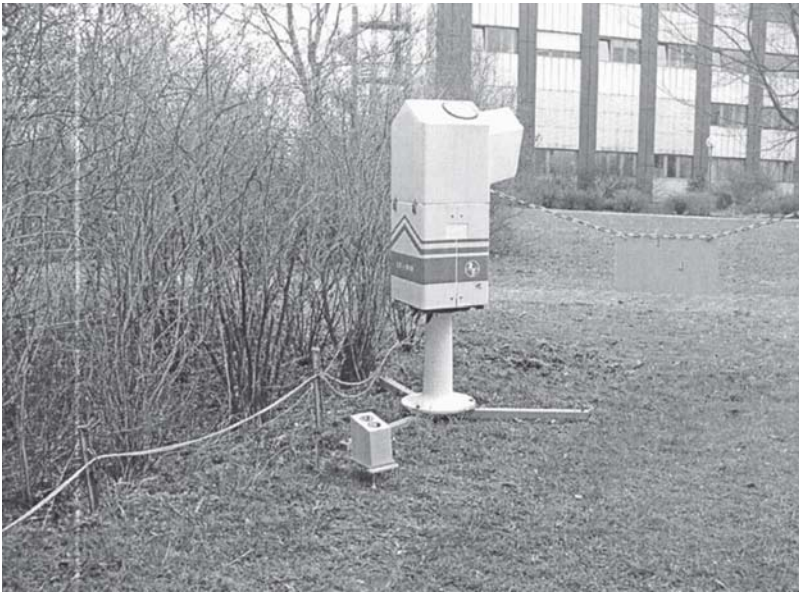


**Fig. 6.4.** Modified Impulsphysik (Vaisala) Ceilometer at Oberpfaffenhofen airstrip (see <http://www.op.dlr.de/ipa/lidar-online>). Photograph also shows (left and in front of Ceilometer) the smaller WHM1k model (cf. Subsection 6.8.2).



every 30 s, although only 10,000 shots per profile are needed. The system is fully automated and can produce horizontal and slant profiles as well, 24 hours a day.

## 6.7 Applications

Range-resolved recording of the backscattered signal allows the depth-resolved measurement of turbidity and, thus, of the local visibility as defined in Eq. (6.1), for distances between several meters and a few kilometers, depending on weather. From that all secondary quantities, whether integrated or not, can be determined. A number of examples is presented below for illustration.

### 6.7.1 Meteorological Optical Range (MOR) at Hamburg Airport

During a campaign at the airport of Hamburg, Germany, in 1991 a visibility lidar was installed near the touchdown point. Figure 6.5 shows 1.5 hours of lidar data along with the results from a standard transmissometer. Although the MOR data varied by more than a factor of 2.5 during the measurement time, the two sets of data are practically identical with, on average, a slight tendency of the lidar to be lower than the transmissometer data (by  $\leq 15\%$ ) and thus on the safe side.

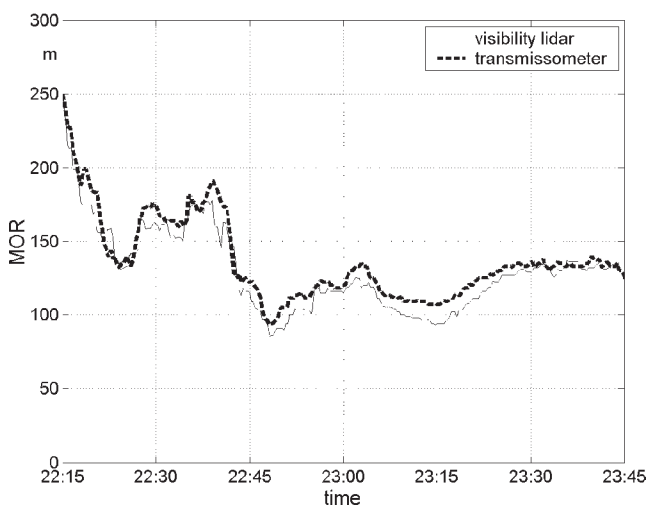


**Fig. 6.5.** Visibility MOR versus time for two sensors, a standard transmissometer and a visibility lidar (airport Hamburg, Germany, 12 January 1992).

### 6.7.2 Slant Visibility (SOR) at Quickborn

Slant visibility SOR (Eq. 6.6) is particularly important for aircraft landing approaches in situations of ground fog and lifted fog layers. A campaign to compare MOR and SOR was staged in Quickborn, Germany, from 1988 to 1990. Two transmissometers, one slant and one horizontally oriented, were used between two masts. A visibility lidar measured from the same position into several elevation angles. The arrangement of the instruments is sketched in Fig. 6.6. Figure 6.7 shows results obtained in three different fog situations. In the event of Fig. 6.7(a) we have relatively homogeneous fog which starts to evaporate around 07:05. Slant visibility SOR increases from about 80 m to more than 1000 m thirty minutes later and is almost identical to the meteorological optical range MOR. In Fig. 6.7(b) a thin layer of fog on the ground affects the horizontal transmissometer, but not the slant instruments which yield much higher visual range values. This is the typical situation in which pilots can see the runway or landing lights but are not allowed to land because the ground transmissometer indicates too dense fog on the ground [3]. Figure 6.7(c) shows the opposite situation in which the fog has lifted from the ground, resulting in good visibility on the ground but poor slant-path visual range [12].

### 6.7.3 Detection of Clouds

Visibility lidars are very well suited for the detection of clouds down to an optical thickness that is hard to perceive with the naked eye from below.

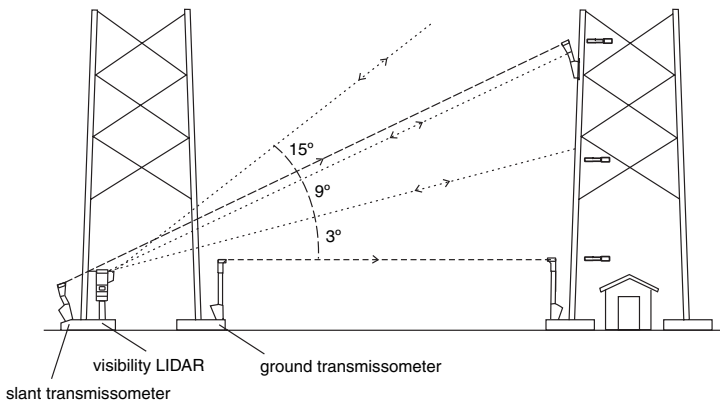


**Fig. 6.6.** Measurement scenario for comparison of horizontal (MOR) and slant visibility (SOR).

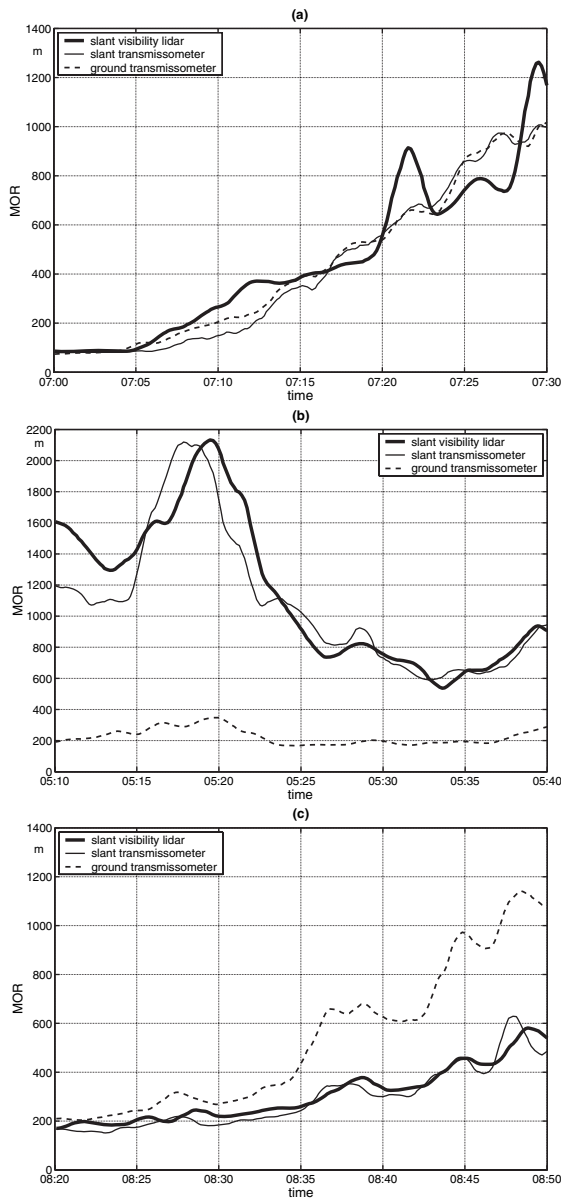


**Fig. 6.7.** Comparison of horizontal (MOR) and slant visibility (SOR). The optical path of the ground transmissometer was at a height of 2 m. The slant transmissometer measured at an elevation angle of  $9^\circ$ . The distance between the transmitter and receiver of the transmissometers was roughly 50 m. The lidar elevation angle was alternated between 3, 9, and  $15^\circ$ . For the comparison the lidar data set was calculated at an elevation of  $9^\circ$ .

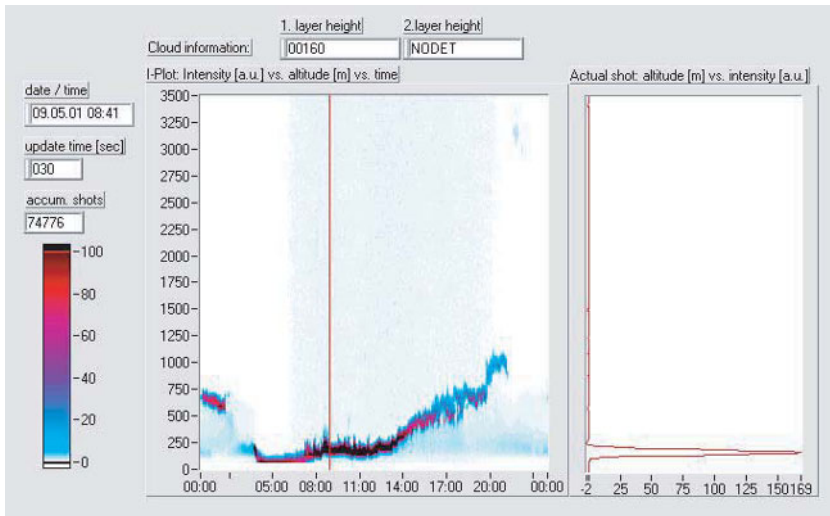


**Fig. 6.8.** Screenshot of vertical fog profile measurements from 8 May 2001, 08:42, to 9 May 2001, 08:41. The plot at right is the actual lidar signal.

Figure 6.8 shows a color-coded intensity plot of the optical density as a function of altitude and time, in a sequence of one profile every 30 s. The red vertical bar is the actual time (08:41 in this case), the data to the right of the red line are the results of the previous day. The seamless transition to the profiles 24 hours before is purely accidental. The actual height profile of the lidar signal is shown on the right. We note the presence of fog and of clouds most of the time, at an altitude that varies between less than 100 m in the early morning and about 1200 m around 20:00 hours. Although the measurement range of the system is 3500 m, the signal gets extinct after 250 m because of the dense fog layer which starts at 160 m altitude.

#### 6.7.4 Cloud Ceiling

Figure 6.9 gives an example of two cloud layers appearing in the profile from a standard commercial ceilometer, illustrating the ability to detect high cirrus clouds.

The standard reporting frequency of ceilometers in use at airports is one set of data every 15 s. An automatic cloud algorithm investigates the shape of the backscatter profile, discards maxima originating from signal noise or falling precipitation, and generates a data message with cloud

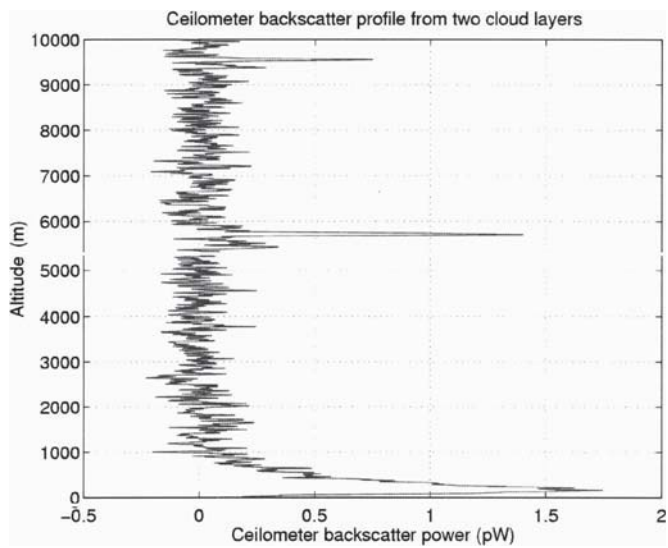


**Fig. 6.9.** Ceilometer backscatter signal from boundary layer aerosol and two cloud layers with base heights of 5700 m and 9600 m. Profile taken with a Vaisala LD-40 Ceilometer on 13 May 2001, 22:15:16 – 22:16:46, averaged over 382,752 pulses.

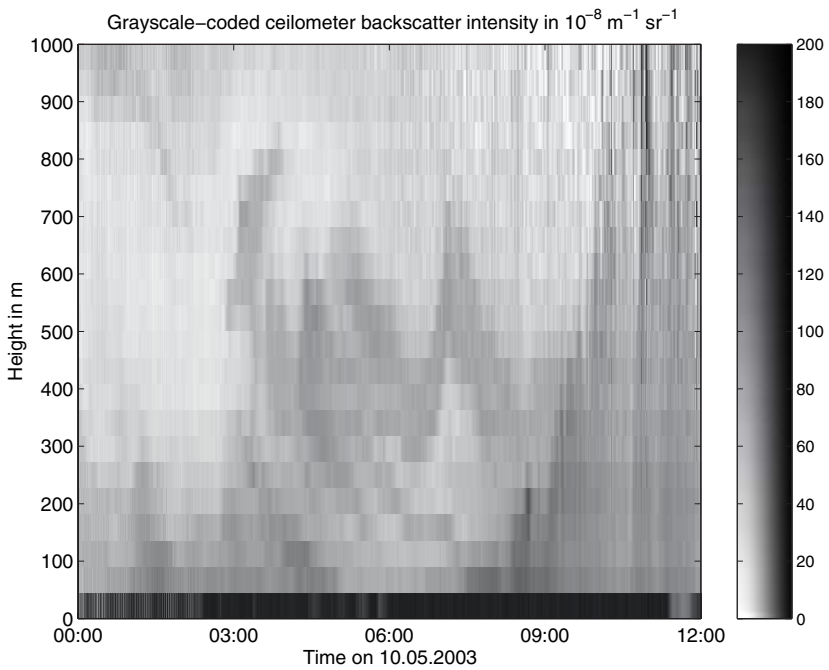


**Fig. 6.10.** Grayscale-coded intensity plot of range-corrected ceilometer backscatter profiles (Hamburg, Germany, 10 May 2003).

base heights and instrument status information. Additional parameters reported include vertical visibility and the amount of precipitation.

Even in clear-atmosphere situations like the one prevailing in Fig. 6.9 there is enough backscatter signal detected from altitudes up to 1000 m to estimate the aerosol concentration in the planetary boundary layer.

### 6.7.5 Mass Concentration Measurements

When the visual range exceeds 2000 m, a standard ceilometer designed to detect cloud bases still receives a considerable amount of backscatter signal from boundary-layer aerosol. The grayscale-coded intensity plot in Fig. 6.10 gives an example.

Comparisons with *in situ* sensors measuring dust concentration values (PM10 and PM2.5) show a good correlation between ceilometer signal and dust concentration measured in the corresponding altitude [13]. Figure 6.11 shows this relationship using an empirically derived

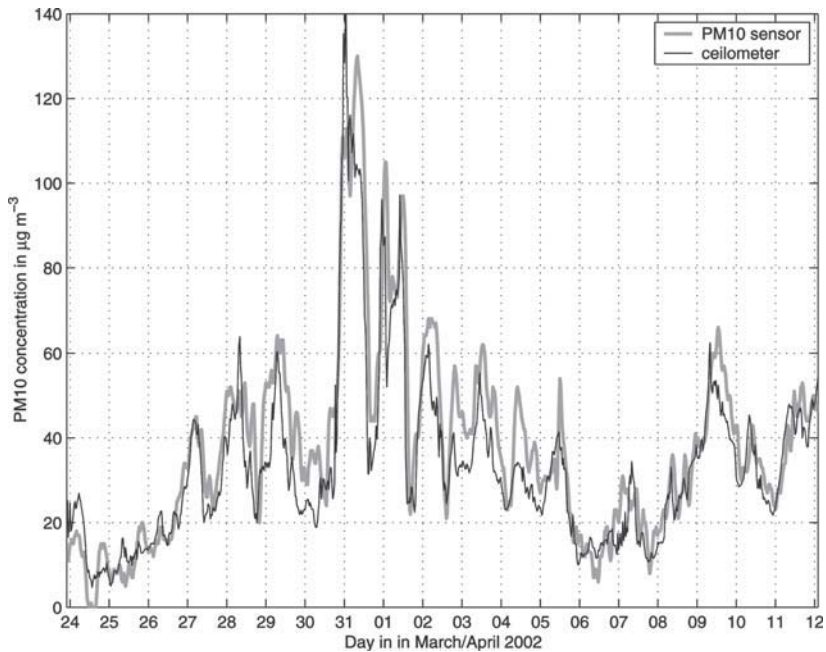


**Fig. 6.11.** Dust concentration derived from ceilometer backscatter between 0 and 30 m altitude and PM10 concentration between 0 and 20 m height (Hannover, Germany, 24 March to 12 April 2002).

linear dependency between ceilometer backscatter and PM10 mass concentration.

## 6.8 Recent Developments

### 6.8.1 Intelligent Taillight: Adaptation of Brightness Using the Lidar Technique

Although in many regions road traffic density has increased dramatically, the rate of accidents has generally decreased. A good deal of this trend is due to the development of equipment that increases traffic safety. The continuation of this process thus deserves particular attention [14, 15].

The perceptibility of automotive lighting and light signals under poor visibility conditions is one important field in this context. The problem is not just precipitation and fog. On a wet road spray whirled up by tires also affects visibility quite strongly. Depending on the amount of moisture on the road and on driving speed, this spray is dragged like a flag more than 20 m behind the vehicle. A measuring principle based on spot-like scanning of only a small volume is not suited for the initialization of any countermeasures. Rather, a method is needed that can measure the turbidity in an extended measuring volume. The exact size of that volume may have to be adapted to the vehicle's speed.

To help improve the visibility of vehicle rear lights to the driver of the vehicle behind, a system is under development for installation in automobile taillights that must be able to detect the following parameters:

- visibility reduction by rain, snow, fog or tire spray,
- distance of the following car and
- speed of the following car.

The sensor data are then transferred to a unit called rear light controller (RLC) in which these and other automotive data are linked to a weather model to generate control signals that regulate the brightness of the lamps. The complete lidar system consists of a transmitter (a laser diode and an optical lens), a receiver (an optical lens and an avalanche photodiode) and a data acquisition system (a digitizer and microcomputer). The whole system including the electronic lamp brightness regulator is housed in a box the size of a car radio. The first prototype is shown in Fig. 6.12 built into the rear part of an automobile. The

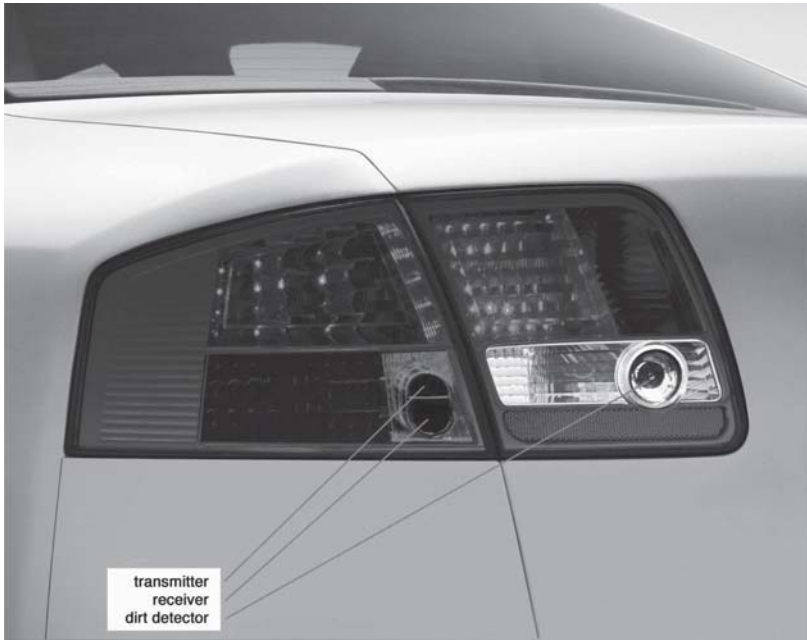


**Fig. 6.12.** Intelligent taillight for automobiles.

integration of the lidar data into a taillight-control computer program and tests in an artificial smoke chamber have been carried out, with very satisfactory results.

### 6.8.2 Miniaturized Visual-Range Lidar for Heliports

In addition to airports where there is generally no serious limitation of space, heliports on boats, drilling platforms, hospital buildings, and the like also need information on cloud base height and visibility. For this purpose a miniaturized visual-range lidar was developed by Jenoptik [16]. The system with model designation WHM1k is considerably more compact than current ceilometers. The difference becomes evident from Fig. 6.4 which shows a WHM1k installed adjacent to an Impulsphysik (Vaisala) standard ceilometer at the DLR site Oberpfaffenhofen.

As can be seen in Fig. 6.13, the two instruments yield essentially the same results. Figure 6.13 also shows that the smaller instrument, which has been designed for shorter operating range, performs very well in situations in which helicopters can still land safely, while safe landing



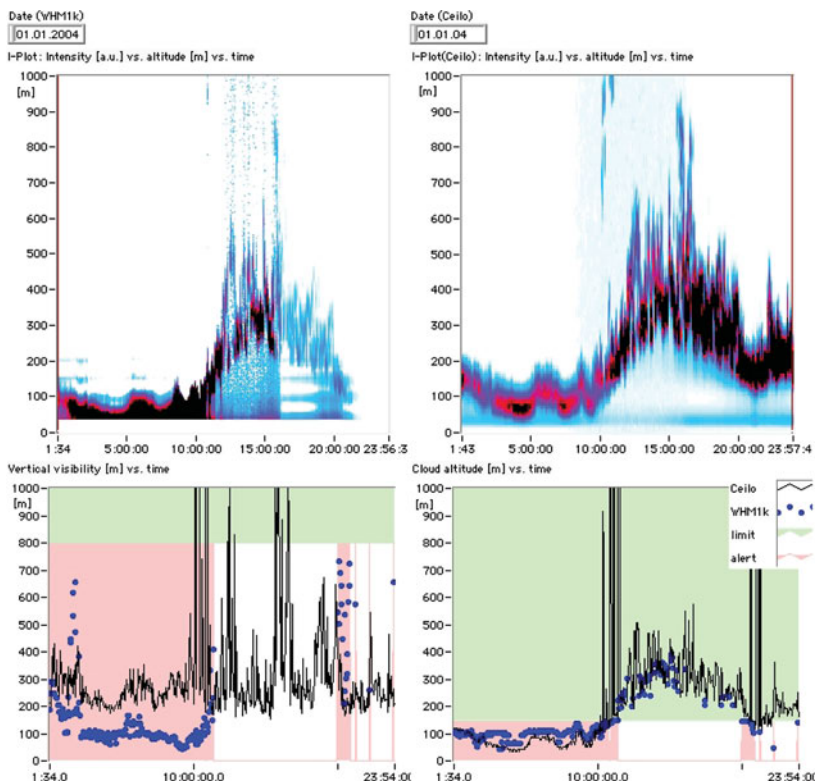


Fig. 6.13. WHM1k optical-density plot from 8 January 2004.

of an airplane would be a considerable challenge. It clearly indicates, however, visibility conditions too poor for a landing approach of the helicopter as well, as is the case between 01:34 and 10:10 in the graph. Figure 6.13 shows the fog recording from 8 January 2004.

Both devices clearly ‘see’ ground fog with patches of varying density from midnight to approximately 11:00. During that time the inversion and cloud detection algorithm registers alarm conditions from the data of both systems: cloud altitude is below 150 m, and vertical visibility is below 800 m. The numerical differences in vertical visibility between the WHM1k (about 100 m) and the ceilometer (about 300 m) are caused by the AC-coupled detection unit of the WHM1k sensor. The smooth signal from the homogeneous fog layer is interpreted as a DC background and cut off by the 1-kHz high-pass filter of the receiver electronics.

From Fig. 6.13 it can also be seen that the fog layer starts to rise at approximately 11:00 hours. The ceilometer still measures some impairment of local visibility, but as this happens at altitudes above 150 m, VFR (i.e., visual flight rule) conditions still prevail. The WHM1k cloud-altitude algorithm follows the rise of the cloud, which seems to become thinner in the WHM1k density plot (top left), but not in the ceilometer plot (top right). This effect occurs only at night. The reason is the fact that the sensitivity control of the APD current is carried out using the background light information. This also needs to be improved. We thus see that downscaling a well-tested, trustworthy, and reliable system still constitutes a technological challenge.

## 6.9 Summary

In summary it can be stated that visibility lidar is an accepted technology wherever impaired vision must be detected to impose speed limits to road or takeoff and landing restrictions to air traffic. Visibility lidars known as ceilometers have reached a degree of maturity to work 24 hours a day in the required fully-automated, hands-off operation mode. The development of much smaller systems for use under restricted space conditions and of systems small and cheap enough to be used as a truck and car accessory is in progress, with good chances to reach full commercial availability soon.

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## Differential-Absorption Lidar for Ozone and Industrial Emissions

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### 7.1 Introduction

During the first two decades of lidar development, much was accomplished with differential-absorption lidar, or DIAL. The basic theory was worked out, the relationship between signal-to-noise ratio and detection limits was elucidated, and DIAL systems in both the ultraviolet (UV) and infrared (IR) spectral regions were developed and fielded for ozone and industrial emissions including  $\text{SO}_2$ ,  $\text{NO}_2$ ,  $\text{NH}_3$ ,  $\text{HCl}$ ,  $\text{CO}$ , hydrazine, and  $\text{Hg}$  [1–3].

In the early days, building lidars was difficult and the technique developed a reputation as something of an arcane art. Even single-wavelength systems were complex and costly and they required highly trained operators and frequent adjustments. These problems were worse for DIAL and so, as promising as the early developments were, DIAL systems saw limited application. For broad acceptance, lidar systems in general needed simpler operation, better reliability, software to produce real-time reduced data, eye safety, standard measurement techniques, and lower costs. DIAL practitioners have made progress in all of these areas during the past two decades, and they have also developed better retrieval techniques and methods for greatly improving measurement accuracy.

In this chapter, the theory of the DIAL technique is reviewed first. Examples of the correction terms are given for the case of a UV ozone lidar, and the considerations for optimizing wavelengths are discussed. Progress in the development of DIAL techniques for ozone

and industrial emissions is then described in four wavelength regions: ultraviolet, visible, mid-infrared, and far infrared. Recent advances in multi-wavelength lidar are summarized, and finally, some conjectures are offered on technology areas that will most likely see rapid progress in the near future.

## 7.2 The DIAL Equation

We begin with the elastic backscatter lidar equation

$$P(R) = P_0 \eta \left( \frac{A}{R^2} \right) \left( \frac{c\tau}{2} \right) \beta(R) \exp \left[ -2 \int_0^R \alpha(r) dr \right] \quad (7.1)$$

where  $P(R)$  is the power received from range  $R$ ,  $P_0$  is the average transmitted power during the laser pulse,  $\eta$  is the receiver efficiency,  $A$  is the receiver area,  $R$  is the range to the scattering volume,  $c$  is the speed of light,  $\tau$  is the laser pulse duration, and  $\beta$  and  $\alpha$  are the atmospheric backscatter coefficient and atmospheric extinction coefficient at range  $R$ .

Next we consider a lidar operating at two wavelengths  $\lambda_{\text{on}}$  and  $\lambda_{\text{off}}$  where a trace gas of interest has correspondingly larger and smaller absorption cross sections, and we define  $P_{\text{on}}$  as the lidar signal at the wavelength  $\lambda_{\text{on}}$  and  $P_{\text{off}}$  as the signal at  $\lambda_{\text{off}}$ . For the purpose of illustration, we assume for the moment that the difference in the atmospheric extinction coefficients at the two wavelengths is solely due to the trace gas, that is,

$$\Delta\alpha = N\Delta\sigma \quad (7.2)$$

where  $N$  is the molecule number density of the trace gas and

$$\Delta\sigma = \sigma(\lambda_{\text{on}}) - \sigma(\lambda_{\text{off}}) \quad (7.3)$$

where  $\sigma$  is the molecular absorption cross section. We also assume that the atmospheric backscatter coefficients at the two wavelengths are identical. In this idealized case, after some algebraic manipulations, we find that

$$N = \frac{1}{2\Delta\sigma} \left[ \frac{d}{dR} \ln \left( \frac{P_{\text{on}}}{P_{\text{off}}} \right) \right]. \quad (7.4)$$

Equation (7.4) shows that DIAL is a self-calibrating measurement technique: all instrument constants are removed by the sequential operations of forming a ratio, finding the logarithm, and taking the derivative

with respect to range. However, a word of caution is in order: the foregoing analysis assumes that there are no range-dependent differences in the lidar responses at the two wavelengths. Fredriksson and Hertz [4] provided an extensive summary of experimental problems that could cause systematic differences at the two wavelengths and consequent errors in the measured value of  $N$ , and Ismail and Browell [5] presented a thorough analysis of the sensitivity of DIAL measurements to both random signal errors and to differential cross section errors. The latter type of error arises from both atmospheric and system effects, including temperature and pressure sensitivities of the trace gas spectrum, Doppler broadening of the Rayleigh return, the non-zero width and a possible shift of the laser line, and uncertainties in its spectral purity and center wavelength. Although the analysis was for a water-vapor DIAL system operating at 720 nm, it can be applied to other gasses and wavelengths.

In practice, lidar signals are not recorded or analyzed as continuous functions, but rather as values in discrete range bins. Expressing the derivative in Eq. (7.4) in terms of a range increment  $\Delta R$ , we have

$$N = \frac{1}{2\Delta\sigma\Delta R} \ln \left( \frac{P_{\text{off}}(R + \Delta R)}{P_{\text{off}}(R)} \frac{P_{\text{on}}(R)}{P_{\text{on}}(R + \Delta R)} \right). \quad (7.5)$$

A real lidar system will have some limit with which it can resolve the term in parentheses in Eq. (7.5), and this fact sets the lidar's *limit of detection*  $N_{\text{LD}}$  for the gas of interest. Assuming that the smallest measurable value of the term in parentheses was 0.02 and using a range increment  $\Delta R$  of 100 m, Collis and Russell [1] derived a table of DIAL detection limits for a combination of 14 gasses and wavelength pairs. Equivalently, Eq. (7.5) can be used in the design of a lidar to find the *minimum range resolution*  $\Delta R_{\text{min}}$ , for a given value of  $\Delta\sigma$ .

Tropospheric DIAL data are usually recorded in fairly small range bins, typically 15 m, and analyzed with a range resolution of 50–300 m. The analysis is not done by simple differencing as in Eq. (7.5) but rather by various curve fitting and filtering techniques that are employed to increase the signal-to-noise ratio. The effect of these techniques on the range resolution actually obtained was studied by Beyerle and McDermid [6].

In the general case, the atmospheric backscatter coefficient is not the same at the two DIAL wavelengths and there is differential extinction due to air molecules, aerosols, and interfering gasses, in addition to the gas of interest. These wavelength-dependent effects require a set of

corrections to Eq. (7.5), as follows:

$$N = \frac{1}{2\Delta\sigma\Delta R} \left[ \ln \left( \frac{P_{\text{off}}(R + \Delta R)}{P_{\text{off}}(R)} \frac{P_{\text{on}}(R)}{P_{\text{on}}(R + \Delta R)} \right) - \ln \left( \frac{\beta_{\text{off}}(R + \Delta R)}{\beta_{\text{off}}(R)} \frac{\beta_{\text{on}}(R)}{\beta_{\text{on}}(R + \Delta R)} \right) \right] - D - E - F \quad (7.6)$$

where

$$D = \frac{\Delta\alpha_{\text{mol}}}{\Delta\sigma} \quad (7.7)$$

is due to the wavelength-dependent extinction of air molecules,

$$E = \frac{\Delta\alpha_{\text{aer}}}{\Delta\sigma} \quad (7.8)$$

is due to the wavelength-dependent extinction of aerosols, and

$$F = \frac{N_{\text{IG}}\Delta\sigma_{\text{IG}}}{\Delta\sigma} \quad (7.9)$$

is due to the wavelength-dependent extinction of an interfering gas. The quantity  $\Delta\sigma_{\text{IG}}/\Delta\sigma$  is sometimes called the *cross sensitivity*  $Q_{\text{IG}}$ .

The molecular number density  $N$ , in units of molecules per  $\text{m}^3$ , is usually converted to a concentration  $C$ , or mass density, by multiplying with the mass  $M$  of one molecule:

$$C = MN. \quad (7.10)$$

$M$  in kg can be found from the molecular weight in atomic mass units (AMU) by using the relation  $1 \text{ AMU} = 1.6605 \times 10^{-27} \text{ kg}$ . A convenient and commonly used unit for concentrations of gaseous pollutants is  $\mu\text{g}/\text{m}^3$ . Sometimes mixing ratios are more practical quantities than concentrations because they remain unchanged when the temperature and pressure change. However, mixing ratios can be by weight or by volume, and this can cause confusion. The former are usually given in kg/kg, g/kg, etc. For the latter, units such as  $\text{m}^3/\text{m}^3$  are not common; instead, percent (%),  $10^{-2}$ , per mill (‰),  $10^{-3}$ , parts per million (ppm,  $10^{-6}$ ), parts per billion (ppb,  $10^{-9}$ ), and parts per trillion (ppt,  $10^{-12}$ ) are used. Although mixing ratios are dimensionless numbers, it must be stated whether the ratios are by weight (as in ppmw) or by volume (as in ppmv), because the numbers are obviously not the same. The volume and mass mixing ratios of a gas with molecular number density

$N$  and concentration  $C$  are simply given by  $N/N_{\text{air}}$ , and  $C/C_{\text{air}}$  with the molecular number density and the density of (dry) air at standard temperature and pressure (STP, 0 °C and  $1.01325 \times 10^5$  Pa) being  $2.687 \times 10^{25}$  molecules/m<sup>3</sup> and 1.2929 kg/m<sup>3</sup>, respectively.

A DIAL system is sensitive to  $N$  or  $C$ , not to the mixing ratio. In order to find a profile in terms of the mixing ratio (such as ppbv), the lidar investigator must have a profile of atmospheric density. Such profiles are often approximated from ground-level pressure and temperature measurements and standard atmospheric lapse rates. This process introduces additional uncertainty into the mixing ratio profile.

The correction terms  $D$ ,  $E$ , and  $F$  in Eqs. (7)–(9) must be subtracted from the first term of Eq. (6). They are independent of the concentration of the gas of interest, and they are generally positive and not negligible. The magnitudes of the corrections can be illustrated with a specific example. We consider a typical UV DIAL system for tropospheric ozone ( $\text{O}_3$ ), with  $\lambda_{\text{on}}$  equal to 288.9 nm and  $\lambda_{\text{off}}$  equal to 299.1 nm. These wavelengths are commonly used for ozone DIAL because they can be conveniently obtained from the fourth harmonic of a Nd:YAG laser (266 nm) by using stimulated Raman scattering (SRS) in high-pressure gas cells containing  $D_2$  and  $H_2$ , respectively. These wavelengths are shown in Fig. 7.1, along with the UV spectrum of ozone. The spectrum of sulfur dioxide ( $\text{SO}_2$ ) is also shown because it is an interfering gas. For this example, we consider the top of the mixing layer, taken to be at 2000 m above

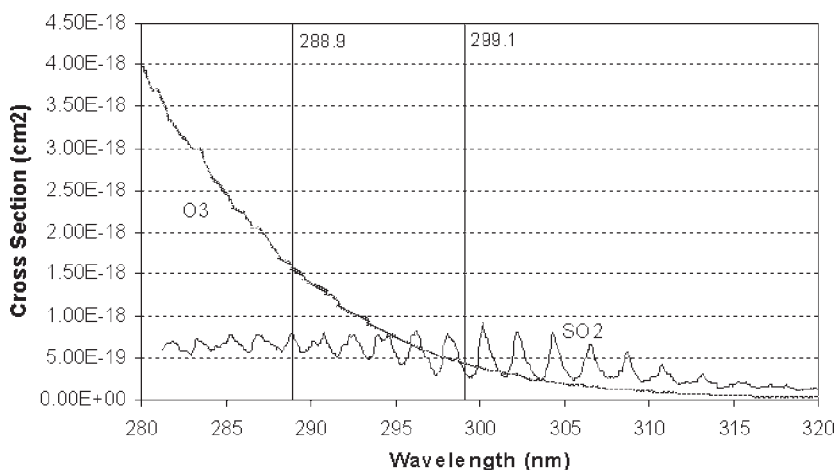


Fig. 7.1. Ultraviolet absorption spectra of ozone and sulfur dioxide.



ground level, where a steep gradient in the aerosol concentration often occurs. The concentration is assumed to decrease by a factor of  $1/e^2$  in a vertical distance of 200 m. The aerosol properties in the mixed layer were calculated by assuming a visibility of 10 km at 550 nm, extinction-to-backscatter ratios  $S_{\text{aer}}$  ranging from 20 to 80 sr, and the Ångström exponent  $a$  ranging from 0 to 1. The variations in the correction terms (caused by variations of  $S_{\text{aer}}$  and  $a$ ) are shown in Table 7.1. These correction terms are not small compared to typical urban daytime ozone values, which are on the order of 50–100 ppbv.

The backscatter gradient term is not simply additive as are terms  $D$ ,  $E$ , and  $F$ . It is due to the difference in the total atmospheric backscatter coefficient at the two wavelengths. It is usually negligible in regions where backscattering is purely molecular in nature, but it becomes large at altitudes where large aerosol gradients exist. This is the primary reason that some DIAL systems employ a third wavelength to independently measure the aerosol profile. However, it should be noted that the early DIAL measurements preceded the development of aerosol inversion techniques that are now commonly used. Sasano, Browell, and Ismail [7] presented a full explanation for the inversion of lidar signals with both aerosol and Rayleigh backscatter in 1985. A useful algorithm for the correction of perturbations by aerosols has also been developed by Goers [8].

The role that aerosol profile inversion techniques play in the accuracy of DIAL results, particularly for the measurement of ozone, has been the topic of many discussions in the literature. Fujimoto, Uchino, and Nagai [9] and Godin et al. [10] systematically compared 4 and 10 different algorithms, respectively. Lidar ozone profiles were also compared with results of *in situ* measurements, showing that carefully taken lidar data differ no more from the results of *in situ* measurement devices than those results vary with respect to one another [11].

The molecular extinction term  $D$  is due to a difference in Rayleigh extinction and, consequently, it can be calculated accurately from the DIAL wavelengths and the air density. For this reason it does not present

**Table 7.1.** DIAL corrections for model atmosphere

| Effect               | Symbol | Correction           |
|----------------------|--------|----------------------|
| Backscatter gradient | —      | 29–39 ppbv           |
| Molecular extinction | $D$    | 7 ppbv               |
| Aerosol extinction   | $E$    | 0 to > 12 ppbv       |
| Interfering gas      | $F$    | $0.4N_{\text{SO}_2}$ |

a major problem for lidar users and researchers. The sign of  $D$  is always positive when  $\lambda_{\text{on}} < \lambda_{\text{off}}$ .

The aerosol extinction term  $E$  is more problematic. As shown by the example summarized in Table 7.1,  $E$  takes values from 0 to 12 ppbv and beyond, depending on the aerosol optical properties, which are generally poorly known. This fact makes the value of  $E$  a large source of uncertainty.

The interfering gas term  $F$  can also be large. For the example shown, the cross sensitivity  $Q_{\text{IG}}$  between  $\text{SO}_2$  and ozone is about 0.4.  $\text{SO}_2$  arises largely from the combustion of fuels, and its concentration depends strongly on the types of fuels in use. In urban areas where high-sulfur coal is used,  $\text{SO}_2$  concentrations on the order of 50 ppbv are not uncommon. This concentration would lead to a +20 ppbv error in the measured ozone concentration, for the values of  $\lambda_{\text{on}}$  and  $\lambda_{\text{off}}$  used in the example.

Proffitt and Langford [12] considered the optimization of  $\lambda_{\text{on}}$  and  $\lambda_{\text{off}}$  in detail for UV DIAL ozone lidar measurements in the free troposphere. The correction terms are small in the free troposphere, where large aerosol gradients are unusual and  $\text{SO}_2$  concentrations are typically on the order of a few ppbv. Unfortunately, the corrections are largest in the mixed layer, where DIAL is a unique tool for measuring pollutants. Minimizing the uncertainties due to the correction terms is therefore especially important in the mixed layer.

As shown in Fig. 7.1, the sulfur dioxide cross section is highly structured in the wavelength region between 277 and 300 nm where most ozone DIAL measurements are made. Weitkamp and others [13] measured the  $\text{SO}_2$  cross section in this range with <3 pm resolution and <4 pm accuracy. They defined 13 pairs of wavelengths, with differences  $\Delta\lambda$  between 0.9 and 8.7 nm, for which the *differential*  $\text{SO}_2$  cross section is zero within  $\pm 3\%$  of each of the respective cross sections. If any of these wavelength pairs is used for ozone DIAL measurements, then the presence of  $\text{SO}_2$  does not cause spurious higher-than-actual  $\text{O}_3$  concentrations. Their wavelength pairs are listed in Table 7.2 in order of decreasing differential ozone cross section. The ozone cross sections are taken from Griggs [14].

We note that the correction terms of Eq. (6) all contain the factor  $1/\Delta\sigma$ , and that they all get larger as  $\Delta\lambda$  becomes larger. It has thus been suggested that the ratio  $\Delta\sigma/\Delta\lambda$  is a figure of merit that should be maximized (although this is just one consideration in choosing the best wavelength pair). For UV ozone lidar, Senff [15], developed the graphical presentation of  $\Delta\sigma/\Delta\lambda$  shown in Fig. 7.2, which shows the relative

**Table 7.2.** Wavelength combinations for ozone DIAL at which the sulfur dioxide differential absorption cross section vanishes

| Wavelength pairs |                            |                             |                                                   | Ozone absorption cross sections                                                                                   |                                                                                                                                          |
|------------------|----------------------------|-----------------------------|---------------------------------------------------|-------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------|
|                  | Signal<br>wavelength       | Reference<br>wavelength     | Wavelength<br>difference                          | O <sub>3</sub> cross<br>section at signal<br>wavelength $\sigma(\lambda_{\text{on}})$ ,<br>$10^{-23} \text{ m}^2$ | Differential O <sub>3</sub><br>cross section<br>$\sigma(\lambda_{\text{on}}) - \sigma(\lambda_{\text{off}})$ ,<br>$10^{-23} \text{ m}^2$ |
| Pair             | $\lambda_{\text{on}}$ , nm | $\lambda_{\text{off}}$ , nm | $\lambda_{\text{off}} - \lambda_{\text{on}}$ , nm |                                                                                                                   |                                                                                                                                          |
| A                | 280.9                      | 289.6                       | 8.7                                               | 35.4                                                                                                              | 20.8                                                                                                                                     |
| B                | 277.6                      | 284.1                       | 6.5                                               | 46.4                                                                                                              | 20.6                                                                                                                                     |
| C                | 280.9                      | 288.3                       | 7.4                                               | 35.4                                                                                                              | 18.3                                                                                                                                     |
| D                | 277.6                      | 282.7                       | 5.1                                               | 46.4                                                                                                              | 16.7                                                                                                                                     |
| E                | 278.6                      | 282.9                       | 4.3                                               | 42.3                                                                                                              | 12.6                                                                                                                                     |
| F                | 284.1                      | 289.6                       | 5.5                                               | 25.8                                                                                                              | 11.2                                                                                                                                     |
| G                | 277.6                      | 280.9                       | 3.3                                               | 46.4                                                                                                              | 11.0                                                                                                                                     |
| H                | 282.9                      | 286.4                       | 3.5                                               | 29.7                                                                                                              | 8.9                                                                                                                                      |
| I                | 286.4                      | 289.6                       | 3.2                                               | 20.8                                                                                                              | 6.2                                                                                                                                      |
| J                | 280.9                      | 282.7                       | 1.8                                               | 35.4                                                                                                              | 5.7                                                                                                                                      |
| K                | 282.7                      | 284.1                       | 1.4                                               | 29.7                                                                                                              | 3.9                                                                                                                                      |
| L                | 286.4                      | 288.3                       | 1.9                                               | 20.8                                                                                                              | 3.7                                                                                                                                      |
| M                | 278.6                      | 279.5                       | 0.9                                               | 42.3                                                                                                              | 2.1                                                                                                                                      |

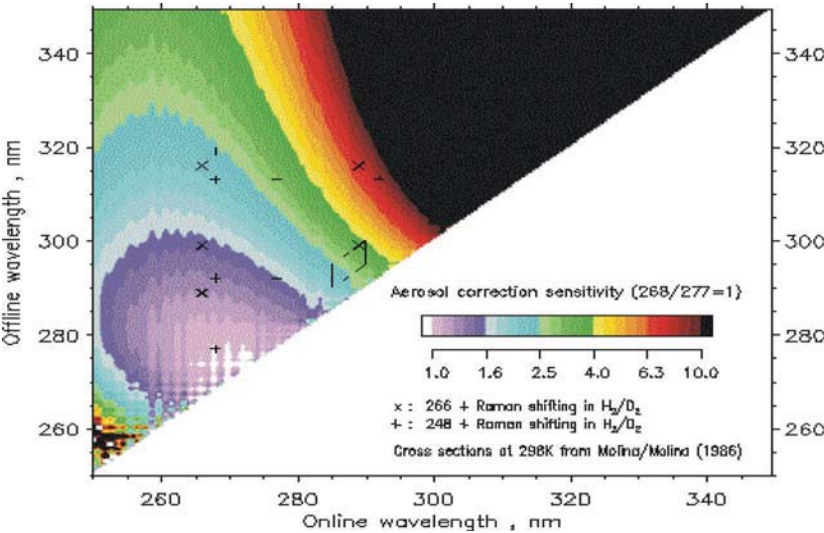


**Fig. 7.2.** Relative sensitivity to aerosol correction for various choices of  $\lambda_{\text{on}}$  and  $\lambda_{\text{off}}$  for UV ozone DIAL [15]. Ozone cross sections taken from Molina and Molina [16]. Crosses (+, x) mark frequently used wavelength combinations.

values of this figure of merit for any choices of  $\lambda_{\text{on}}$  and  $\lambda_{\text{off}}$  between 250 and 350 nm. Note that *low* aerosol correction sensitivity represents a *large* figure of merit.

In Fig. 7.2 six popular wavelength combinations have been marked with crosses. Two of them are based on the krypton fluoride laser with the 268-nm deuterium first Stokes-shifted line as  $\lambda_{\text{on}}$  and the hydrogen first and second Stokes-shifted lines as  $\lambda_{\text{off}}$  (+), four use the fourth harmonic of a Nd:YAG laser, both unshifted (266 nm) and first-Stokes-shifted in  $D_2$  (289 nm), as  $\lambda_{\text{on}}$  and several Stokes-shifted lines from  $H_2$  and  $D_2$  as  $\lambda_{\text{off}}$ , ( $\times$ ). Ozone lidars have been reported that use for  $\lambda_{\text{off}}$  wavelengths as high as 316 nm, obtained from a second-Stokes Raman shift in  $D_2$ , but consideration of  $\Delta\sigma/\Delta\lambda$  shows that such a choice of wavelengths is not a good practice. An appropriate value of  $\Delta\sigma$  can be obtained, but the figure of merit is decreased substantially compared to shorter wavelengths, which means that the correction terms, and their uncertainties, are much larger.

Optimizing the choices of  $\lambda_{\text{on}}$  and  $\lambda_{\text{off}}$  for a given application requires a specification of the maximum range and the range resolution required, the spectra of the trace gas and any interfering gasses, and the expected concentrations of the trace gas and the interfering gasses. Wavelength optimization for UV ozone DIAL is achieved through the following considerations:

1. The ozone cross section at the wavelength  $\lambda_{\text{on}}$  must be low enough that the lidar can meet its maximum-range requirement.
2. The wavelengths  $\lambda_{\text{on}}$  and  $\lambda_{\text{off}}$  must be chosen so that  $\Delta\sigma$  is large enough to meet the range resolution requirement [Eq. (5)].
3. The ratio  $\Delta\sigma/\Delta\lambda$  should be maximized.
4. The wavelengths  $\lambda_{\text{on}}$  and  $\lambda_{\text{off}}$  should be chosen to minimize  $\Delta\sigma_{\text{IG}}$ .

As an example, an ozone DIAL system with a maximum range of 3 km and a range resolution of 200 m, for ozone concentrations up to 100 ppbv and  $\text{SO}_2$  up to 50 ppbv, would have  $\lambda_{\text{on}}$  and  $\lambda_{\text{off}}$  in the 285 to 295 nm range with  $\Delta\lambda$  about 5 nm. As mentioned above,  $\Delta\sigma_{\text{IG}}$  can be made negligibly small by choosing a wavelength pair from Table 7.2.

In another case in which a shorter maximum range could be tolerated but higher  $\text{O}_3$  concentrations had to be accommodated, the optimization led to the choice of  $\lambda_{\text{on}} = 280.91$  nm and  $\lambda_{\text{off}} = 282.72$  nm, i.e., wavelength pair *J* of Table 7.2. The individual steps of this optimization are described in detail in [8].

## 7.3 DIAL Systems

### 7.3.1 Ultraviolet DIAL Systems

The ultraviolet (UV) spectral region is useful for DIAL studies of pollution and industrial emissions because several pollutants have spectra in the UV region, because eye-safety requirements can be met in the UV, and because the sky background radiance is small, which makes daytime operation possible.

Ultraviolet DIAL systems have been developed and fielded in several countries around the world during the past two decades. Some of these lidars were in mobile vans, and the vans with the most published activity were developed in Sweden, first at the Chalmers Institute of Technology [17] and later at the Lund Institute of Technology [18]. All of the early Swedish mobile lidars were based on two Nd:YAG-pumped dye lasers, and all had a scanning mirror for mapping pollutant concentrations in horizontal or vertical slices.

In 1984, Swedish investigators reported a study of  $\text{SO}_2$  from a paper mill that included concentration maps as well as an estimate of the flux of  $\text{SO}_2$  from the mill [19], and in 1987 they mapped out  $\text{NO}_2$  during an inversion episode that led to a particularly high ground-level  $\text{NO}_2$  concentration [20].

Edner and others [20] also reported measurements of molecular chlorine,  $\text{Cl}_2$ , using DIAL wavelengths in the 298–308 nm region. The UV spectrum of  $\text{Cl}_2$  is featureless, so the problems involved in choosing DIAL wavelengths are somewhat like those for  $\text{O}_3$  except that the cross section differences obtainable are about two orders of magnitude smaller for  $\text{Cl}_2$ . For this reason, the authors were only able to detect strong artificial sources of  $\text{Cl}_2$ . They also pointed out that their expected sensitivity using a 10-nm wavelength separation would be about seven times worse than that for  $\text{NO}_2$  and that other gasses, such as  $\text{O}_3$  and  $\text{SO}_2$ , would be strong interferents for open-atmosphere measurements. For these reasons, it is difficult to envision a practical application of UV DIAL for chlorine.

In 1989, the Swedish researchers reported DIAL monitoring of elemental mercury, Hg, pointing out that mercury is an unusual atmospheric pollutant because it occurs in elemental form [21]. Previous attempts had been made, but sensitive measurements had to await the availability of a high-power narrow-band laser at 254 nm, which was equipped with a dual-wavelength feature, producing on- and off-line pulses alternately. Mercury DIAL is complicated by the occurrence of

resonance fluorescence, so data processing must include some subtle differences from ordinary DIAL. The system was tested on plumes from a chlorine-alkali plant where mercury was used in the processing. Using the scanning DIAL system, the investigators mapped out the Hg plume and, with wind speed data, found the flux, which was 30 g/hr of atomic Hg. With a range resolution of 1 km, the investigators could measure concentrations on the order of  $1 \text{ ng/m}^3$ , which compares favorably with the average European background level of  $1.6 \text{ ng/m}^3$ .

Extensive improvements to the mobile van were reported in 2003 [22]. The lidar transmitter system is now all solid state, with two injection-seeded Nd:YAG lasers pumping optical parametric oscillators (OPOs) along with several harmonic generators to cover the wavelength range from 220 nm to about  $4 \mu\text{m}$ . The van also incorporates instrumentation for laser-induced fluorescence (LIF) measurements on both aquatic and terrestrial targets.

Investigators in Germany developed a mobile UV DIAL van known as ARGOS (Advanced Remote Gaseous Oxides Sensor) [23]. ARGOS was designed to measure  $\text{O}_3$ ,  $\text{NO}_2$ , and  $\text{SO}_2$ . The lidar is based on two Nd:YAG-pumped dye lasers, and the optical system includes a two-axis scanner. The ARGOS van also has a three-component Doppler sodar (sound detection and ranging) that measures the vertical profile of the wind vector for emission measurements and for investigations of transport phenomena in general. The ARGOS system is shown schematically in Fig. 7.3. The on and off wavelengths used are 296.17/297.35 nm for  $\text{SO}_2$  and 280.92/282.72 nm for  $\text{O}_3$ . The rationale for these wavelength choices was described by Weitkamp and others [13].

Goers [24] described measurements of  $\text{SO}_2$  and  $\text{O}_3$  with ARGOS. The  $\text{SO}_2$  measurements included emissions from a zinc smelter in Germany and steel mills in Brazil. As described earlier, DIAL measurements must be corrected for several error terms. An exceptionally detailed account of DIAL data processing, including aerosol correction algorithms, was given by Goers in her PhD thesis [8].

In the United States, a mobile DIAL van for use in air quality studies was constructed by the Environmental Technology Laboratory (ETL) of the National Oceanic and Atmospheric Administration (NOAA) [25–27]. The system, known as OPAL (Ozone Profiling Atmospheric Lidar), is quite different from the European vans because it was designed to obtain vertical distributions of one gas only,  $\text{O}_3$ . The lidar uses harmonics of Nd:YAG, Raman shifting with  $\text{H}_2$  and  $\text{D}_2$  (with both gasses in the same cell), and sum frequency mixing to generate 266, 289, 299, and

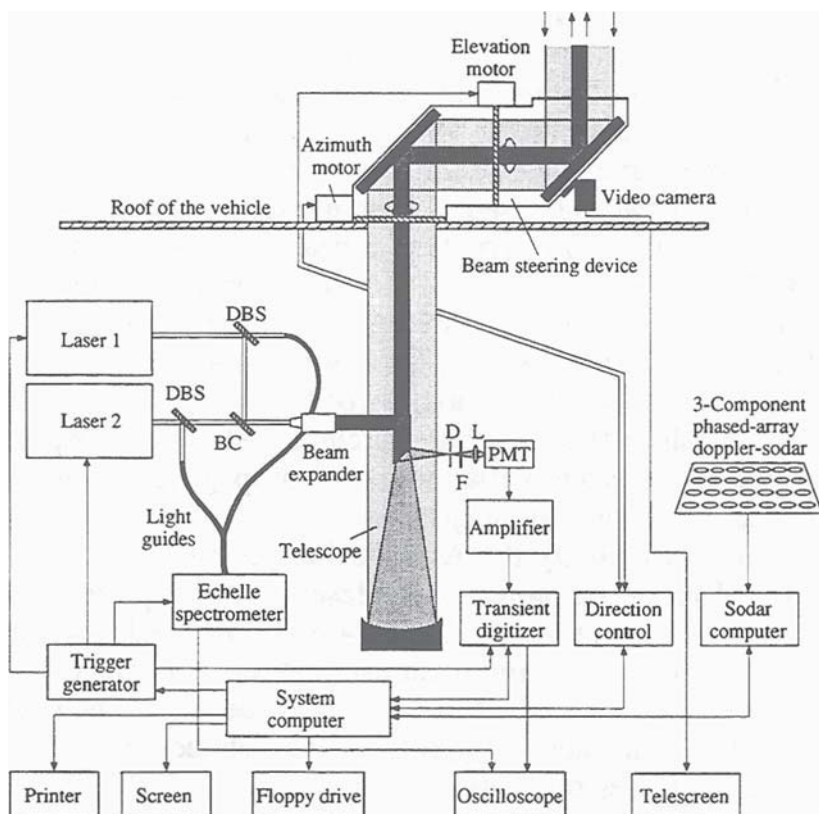


**Fig. 7.3.** The ARGOS system: DBS, dichroic beamsplitter; BC, polarizing beam combiner; D, diaphragm; F, filters; L, lens; PMT, photomultiplier tube [23].

355 nm. All wavelengths are transmitted simultaneously and separated in the receiver with filters and a spectrometer. The 355-nm signal is used to measure the aerosol profile. OPAL has been used in many air quality campaigns over the past decade, with continuous upgrades and improvements in its optical configuration.

A European system that, like OPAL, is exclusively for the measurement of ozone vertical profiles also employs Raman shifting, but it is based on a KrF excimer laser. The wavelength shifting occurs in one Raman cell only, and the first and second Stokes Raman lines of  $H_2$  at 277.2 and 313.2 nm are used as the on- and off-resonance wavelengths [28].

Most European DIAL vans employ dye lasers for tunability whereas the NOAA OPAL system uses Raman shifting for compactness, simplicity and low maintenance effort. Ideally, a laser source would provide all of these attributes. The OPO is a compact, tunable, solid-state device that is starting to appear in lidar systems. In 2002, Fix and others [29] reported an extensive investigation of OPO sources for tunable UV radiation, using several different harmonics of Nd:YAG for both pumping the OPO cavity and for sum frequency mixing. These investigators obtained pulse energies up to 10 mJ in the 281–291 nm region and 4% conversion efficiency from the Nd:YAG fundamental at 1064 nm by utilizing intracavity mixing, as shown in Fig. 7.4. They enumerated the requirements of the OPO source for DIAL, including a robust design, easy handling, low cost, tunability, narrow linewidth, and wavelength stability. A lower-power version of their OPO source is used in the Ozone Profiler marketed by Elight Laser Systems of Teltow, Germany. The Ozone Profiler is an unattended system with an altitude range from 100 m up to 2 km.

Another O<sub>3</sub> DIAL system based on OPOs was reported by Zenker and others [30], who developed tunable OPO sources capable of transmitting as much as 25 mJ per pulse in the ultraviolet. The Swedish van also now uses OPOs, as mentioned above.

Lidar researchers at the Georgia Tech Research Institute in Atlanta, Georgia, are also developing an unattended O<sub>3</sub> DIAL system, in a partnership with LaserCraft, Inc. of Norcross, Georgia [31,32]. The Georgia

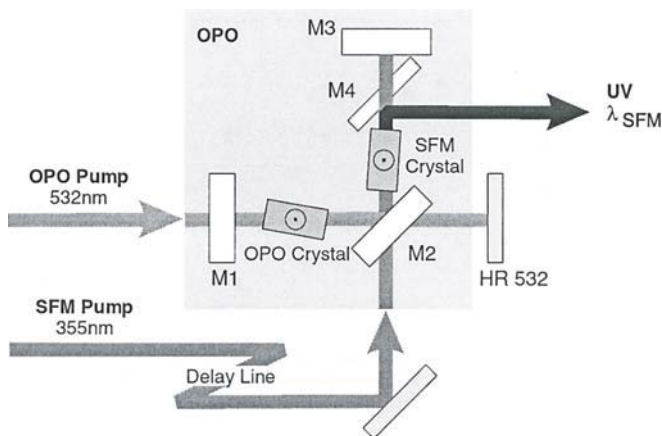


**Fig. 7.4.** Basic setup of the optical parametric oscillator (OPO) with intracavity sum-frequency mixing (SFM) [29].



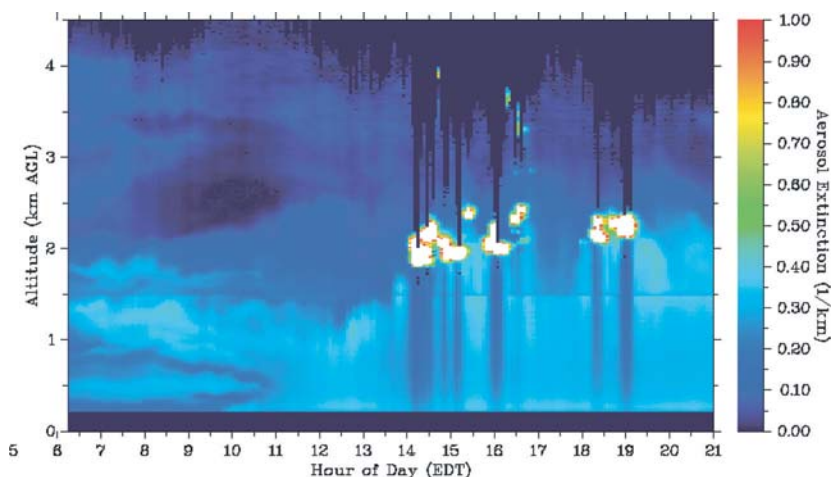


Fig. 7.5. Aerosol extinction profiles produced in real time by NEXLASER.

Tech system, known as NEXLASER (NEXt generation Laser Atmospheric sensor for the SouthEastern Region), is designed specifically for polluted urban environments. It has a maximum altitude of 3 km. NEXLASER is intended for deployment in networks of multiple units, providing real-time ozone profiles throughout an urban area to a central site via the Internet. The first version of the NEXLASER lidar was based on the NOAA OPAL system, using Raman shifting in  $H_2$  and  $D_2$  to obtain the DIAL wavelengths, which are susceptible to  $SO_2$  as an interfering gas. Many urban environments have high concentrations of  $SO_2$ ; for this reason, an OPO source is currently being investigated, along with other Raman-shifting gasses. NEXLASER features unattended operation and real-time reduced data, including both ozone profiles and aerosol extinction coefficient profiles. Examples are shown in Figs. 7.5 and 7.6.

### 7.3.2 Visible-Light DIAL Systems

The only application of visible-light DIAL to industrial emissions has been to nitrogen dioxide ( $NO_2$ ), which has a spectrum with maximum cross section differences around 440 nm, as shown in Fig. 7.7.  $NO_2$  is produced by combustion processes, and it is also the direct precursor of ozone.

Early estimates of the measurement precision of  $NO_2$  lidars by Collis and Russell [1] and by Takeuchi and others [34] were not encouraging except for plumes, being in the range 100–1000 ppbv. However, the

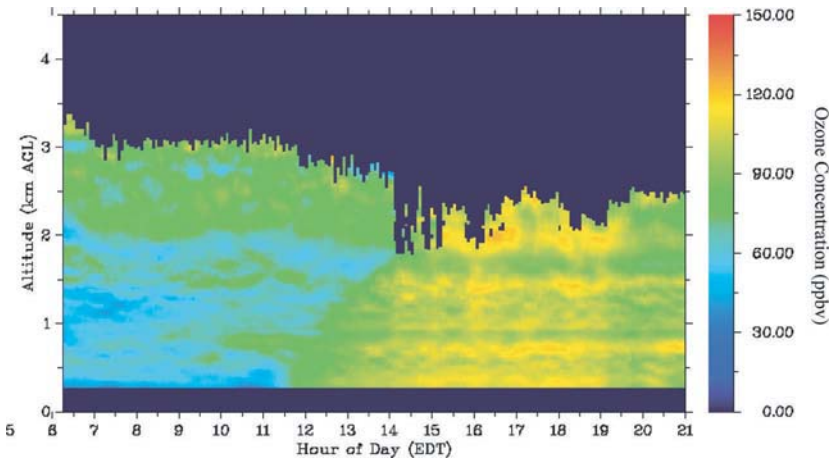


**Fig. 7.6.** Ozone profiles produced in real time by NEXLASER.

mobile DIAL system developed in Sweden [17] that used wavelengths near 448 nm was thoroughly evaluated by Fredriksson and Hertz [4], who found its measurement capabilities to be much better than the early estimates. Staehr and others [35] later demonstrated the detection of  $\text{NO}_2$  to levels of 10 ppbv at ranges up to 6 km, with a two-laser system operating near 450 nm with near-simultaneous pulses at the two wavelengths. These authors also used what has come to be known as *null profiles* (recorded with both lasers tuned to the same wavelength) to determine their measurement uncertainties.

Kölsch and others [36] developed a concept in 1989 for simultaneously measuring NO and  $\text{NO}_2$  with a pair of wavelengths near 454 nm

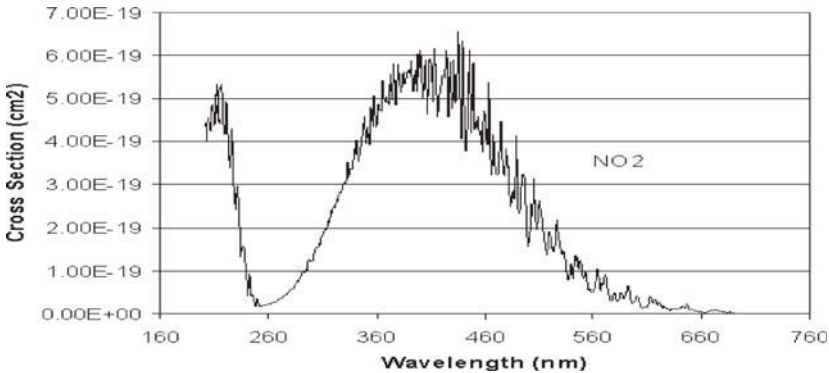


**Fig. 7.7.** The spectrum of  $\text{NO}_2$  (from W. Schneider et al. [33]).

frequency-doubled to a pair near 227 nm. They demonstrated their idea with field measurements, using an excimer-pumped dye laser. In 1990 and 1991, Swart and other investigators reported a mobile NO<sub>2</sub> lidar in the Netherlands applied to emissions from stacks and freeway traffic [37, 38]. A mobile system was also built by Fritzsche and Schubert to simultaneously measure either two of the gases SO<sub>2</sub>, NO<sub>2</sub>, and O<sub>3</sub> or else one of those gases along with meteorological visual range [39].

Toriumi and others [40] reported an NO<sub>2</sub> lidar in 1996 that was based on a titanium:sapphire (Ti:Al<sub>2</sub>O<sub>3</sub>) laser operating at 447.9 nm (on) and 447.2 nm (off). The lidar, which was used to measure the exhaust plume from a diesel generator at a range of 125 m, had a range resolution of 12 m and a detection limit of 200 ppbv.

Yu and others [41] reported on a dual-wavelength Ti:sapphire laser and considered its use for NO<sub>2</sub> lidar, with on and off wavelengths of 398.3 and 397.5 nm, respectively. These wavelengths are just below the eye-safety cutoff at 400 nm, making eye-safe operation orders of magnitude easier to achieve. The transition to eye-safe operation may make NO<sub>2</sub> DIAL much more useful, especially in urban areas where it is difficult to ensure that personnel will never encounter the laser beam.

In Japan, Nayuki and others [42] recently developed an NO<sub>2</sub> lidar using sum frequency generation of 448-nm light with an Nd:YAG-pumped dye laser, and they reported measurements of concentrations in the range 10–50 ppbv at altitudes of 1–2 km, at night. They estimated their experimental error as  $\pm 7$  ppbv. The same authors later reported a DIAL system for simultaneous O<sub>3</sub> and NO<sub>2</sub> measurements with a measurement uncertainty of 5.5 ppbv in the 1.0–1.5 km altitude range [43]. These measurements were also made at night, because the sky background makes daytime measurements impractical.

Currently, eye-safe NO<sub>2</sub> lidars based on Ti:Al<sub>2</sub>O<sub>3</sub> lasers are in use in Europe, and NO<sub>2</sub> lidars based on dye lasers are under development in Japan. Results to date show that measurement uncertainties on the order of 5 ppbv can be achieved at ranges of several km with range resolutions on the order of 100 m. Current eye-safe NO<sub>2</sub> DIAL systems are therefore useful for routine monitoring in polluted urban environments, where daytime NO<sub>2</sub> concentrations as high as 50 ppbv are not unusual.

### 7.3.3 Mid-Infrared DIAL Systems

Mid-IR DIAL measurements were made as early as 1980 by Weitkamp and others [44], who measured concentrations of hydrogen chloride

on the order of 1 ppm in the plume from a shipborne incinerator, by using a deuterium-fluoride (DF) laser to generate lines near  $3.6\text{ }\mu\text{m}$ . In 1987, Menyuk and Killinger [45] reported a mid-IR lidar based on a Co:MgF<sub>2</sub> laser that was tunable from 1.5 to  $2.3\text{ }\mu\text{m}$ , with a pulse energy of 10 mJ and 3 Hz pulse repetition frequency (PRF). The lidar receiver was based on a 0.3-m telescope with a cooled InSb detector. Extrapolating from measurements of HCl in a semi-enclosed cell at a concentration of 40 ppm, these investigators estimated that their lidar could measure HCl at concentrations of a few ppm at ranges of 2 km, and that the detection sensitivity for CH<sub>4</sub> would be an order of magnitude worse.

Milton and others [46] measured a range of organic and inorganic species including methane, propane and butane, along with other gasses amenable to lidar determinations in the UV and visible. They mixed radiation in the 785–851 nm region from a tunable dye laser with the output of a Nd:YAG laser to obtain IR radiation between 3.0 and  $4.2\text{ }\mu\text{m}$ , which was then amplified in a pulsed optical parametric amplifier (OPA) [47].

Uchiumi and others [48] reported a mid-IR lidar based on a Raman-shifted Ti:Al<sub>2</sub>O<sub>3</sub> laser, tunable from 680 nm to  $3.2\text{ }\mu\text{m}$ . These authors found optimum wavelength pairs and calculated the maximum detection ranges for CH<sub>4</sub>, CO<sub>2</sub>, CO, and N<sub>2</sub>O, finding that the maximum altitude ranges should be 2–3 km for all four gasses.

In 2000, Ambrico and others [49] published an extensive sensitivity analysis for mid-IR DIAL that included the following gasses: HCl, CO, CO<sub>2</sub>, NO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>O, and O<sub>2</sub> (as a reference gas). A wealth of DIAL line pairs was considered for three scenarios: clean air, urban polluted air, and stack emissions. The authors presented arguments that the mid-IR is a good wavelength region for DIAL measurements, particularly because tunable laser sources based on OPOs are now becoming available.

Despite the optimistic predictions of several investigators, few mid-IR DIAL measurements have been made to date. It is not yet clear whether routine DIAL measurements will eventually be made in the mid-IR.

### 7.3.4 Far-Infrared DIAL Systems

Quagliano and others [50] listed several advantages of the far-infrared region (8–12  $\mu\text{m}$ ) for gas detection, including the availability of CO<sub>2</sub> lasers that are line-tunable over much of this spectral region, the fact that many industrial gasses have unique far-IR spectra, and transparency of the atmosphere at these wavelengths. The authors investigated the

detection of trichloroethane, trichloroethylene, Freon 113, and ethylene, but their system measured only path-integrated concentrations because it relied on hard target returns. Ahlberg, Lundqvist, and Olsson [51] also described long-path measurements of ethylene using a CO<sub>2</sub> laser.

Force and others [52] reported laser remote sensing of ammonia (NH<sub>3</sub>) with a CO<sub>2</sub> laser system, again using topographic targets. Their technique used the 10P(30) line at 10.69 μm and the 10P(32) line at 10.71 μm as the off and on lines, respectively. They were able to measure NH<sub>3</sub> concentrations from 5 to 20 ppbv with 30-s averaging. They also used the R(30) line at 9.22 μm and the R(26) line at 9.24 μm to measure NH<sub>3</sub> concentrations of  $15 \pm 5$  ppbv by using a retroreflector at 2.7 km range.

In 1995, Carlisle and others [53] reported both hard-target and range-resolved measurements of chemical-vapor plumes using a CO<sub>2</sub>-laser based DIAL system known as ADEDIS (from the French phrase *appareil de détection à distance*). They were concerned with the organophosphates triethylphosphate (TEP), diethylmethylphosphonate (DEMP), and di-isopropylmethylphosphonate (DIMP). They were able to measure TEP with a standard deviation of 0.13 mg/m<sup>3</sup> at a range of 1 km.

Barbini and others [54] described a dual-wavelength CO<sub>2</sub> laser for DIAL measurements and presented horizontal measurements of both O<sub>3</sub> and H<sub>2</sub>O to a range of about 1 km. Their laser system transmitted about 300 mJ per pulse.

Although CO<sub>2</sub> lasers can provide numerous lines in the 9–11 μm region, the electronic noise associated with currently available detector/preamplifier combinations for direct detection is high and the atmospheric backscatter coefficient is generally very small. For these reasons, direct detection lidar in the far IR is insensitive and DIAL measurements are limited to ranges of about 1 km or less.

A 10-μm lidar can, of course, be made much more sensitive by using coherent detection. In 2002, Zhao and others [55] reported DIAL measurements with a coherent Doppler lidar in a field experiment with controlled releases of NH<sub>3</sub> plumes. The lidar was used to measure both wind speeds and NH<sub>3</sub> concentrations in the plumes, from which the investigators derived fluxes. Their measured fluxes were within 10% of the values calculated from the release rates, showing that their lidar technique can be used to find fluxes with a useful accuracy. Their measurement geometry and flux comparisons are shown in Figs. 7.8 and 7.9. Although the concentrations in the plume were rather large, Zhao [56] has shown that a different line pair could be used to measure

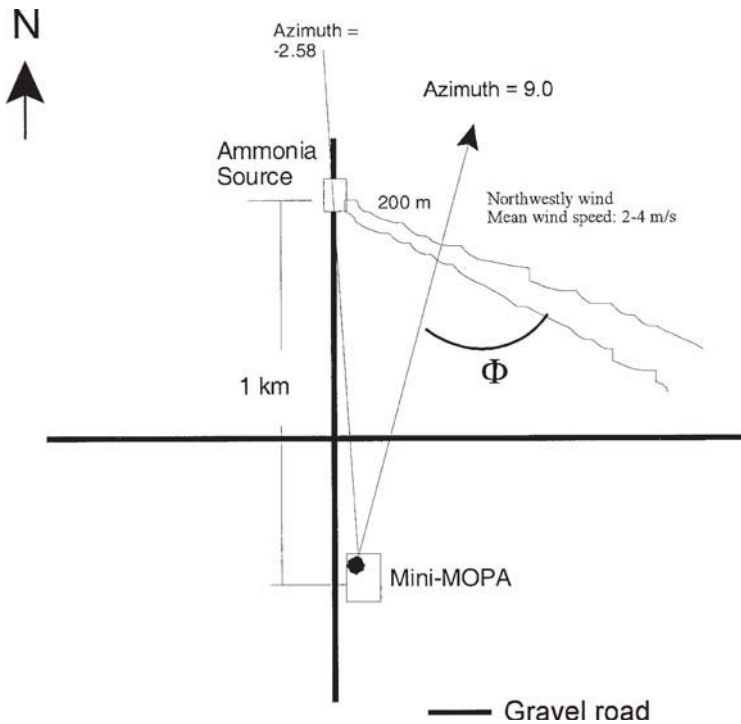


Fig. 7.8. Locations of the lidar and ammonia source and plume [55].

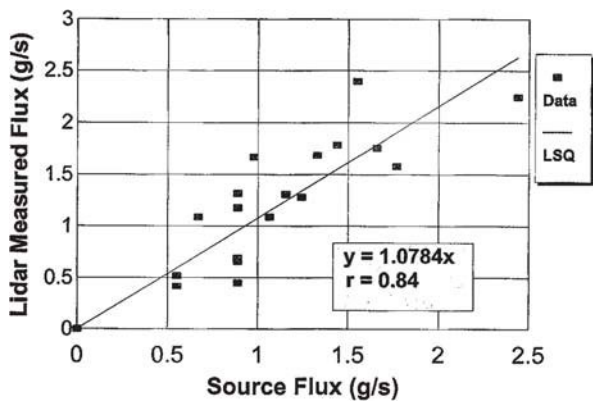


Fig. 7.9. Comparison of source fluxes with lidar-measured fluxes [55].

three-dimensional concentrations in the 5–100 ppbv range in an area 5 km on a side.

## 7.4 Multi-Wavelength DIAL

The DIAL technique can be viewed as a spectroscopic analysis reduced to its absolute minimum number of spectral elements, in which a gas concentration is determined from measurements at just two wavelengths, on and off the maximum of a spectral feature. By contrast, spectrometers commonly used in chemical analysis and in atmospheric studies employ thousands of spectral resolution elements to determine the concentrations of multiple gasses with overlapping spectra. Using more than two wavelengths is an obvious extension of the DIAL technique, yet multiple wavelengths have not been employed until recently, with the exception of line-tunable CO<sub>2</sub> laser systems using topographic targets. In addition to measuring more than one gas at a time, multiple wavelengths may also provide better knowledge of aerosol optical properties, which would improve the accuracy of aerosol corrections. New optimization schemes and new analysis algorithms will be needed for choosing the optimum wavelengths and to make full use of the additional data. The lidar community has just started to explore the advantages of multiple-wavelength systems.

Several different approaches to multiwavelength DIAL have been reported, including three-wavelength ozone lidar techniques to eliminate aerosol corrections and SO<sub>2</sub> interference, DIAL systems in which two gases are measured separately, three- and four-wavelength systems to provide immunity to an interfering gas or to otherwise improve measurement accuracy, and multiwavelength systems that measure two or more gasses simultaneously.

In 1985, Fredriksson [57] discussed the potential advantages of multiwavelength DIAL in connection with planned upgrades to the Swedish mobile lidar van, and the selection of a set of seven wavelengths in the mid-IR for the simultaneous measurement of methane, ethane, and propane was recently discussed by Weibring and others [22]. Warren [58] investigated the concept of using multiple wavelengths to measure multiple gasses by developing a methodology for estimating the detection and discrimination performance of a multiwavelength DIAL. Warren considered column content only, for two gasses. He found that a full matrix of uncertainty values was necessary because of correlations

in the statistical fluctuations of column content values, and pointed out that optimum wavelength values must be chosen with the matrix in mind.

Three-wavelength ozone measurements were proposed in 1994 and 1996 by Chinese investigators Wang and others [59, 60] who presented a theoretical analysis and a numerical simulation of a technique they termed *Dual DIAL*, proposing to use two pairs of lines that were obtainable from single-frequency lasers by Raman shifting. Their analysis showed that ozone measurements could be made much less sensitive to aerosol effects and also nearly immune to interference from  $\text{SO}_2$  and  $\text{NO}_2$  by using three wavelengths. Kovalev and Bristow [61] referred to Dual DIAL as a *compensational* technique, and discussed a three-wavelength version of it in which no correction is needed for aerosol differential extinction and backscattering. They also pointed out that errors due to interfering species could be decreased by a proper choice of wavelengths.

Rambaldi and others [62] reported five-wavelength DIAL measurements of  $\text{SO}_2$  and  $\text{O}_3$  using a system based on a Ce:LiSAF laser that was tunable from 284 to 299 nm. Their analysis was based on finding the  $\text{SO}_2$  concentration first and then using it to correct the  $\text{O}_3$  measurements, because they were measuring  $\text{SO}_2$  concentrations as high as 1 ppmv whereas the  $\text{O}_3$  concentration was only about 15 ppbv.

Strong and Jones [63] presented a theoretical analysis in 1995 for a novel instrument reported earlier by Jones [64]. The instrument employed a broadband, pulsed source (a xenon flash lamp), a spectrometer, and a CCD camera in order to generate range-resolved spectra. The authors used formal retrieval theory to estimate the errors in simultaneous profiles of  $\text{O}_3$ ,  $\text{H}_2\text{O}$ , and  $\text{NO}_2$ , giving special attention to the effects of the flashlamp pulse duration and the CCD recording interval. They concluded that it should be possible to obtain vertical profiles of all three gasses simultaneously with 30% uncertainty up to altitudes of 12–15 km with a 3-km range resolution, using an integration time of 20 min. Apparently the technique is intended only for use in total darkness.

The most recent multiwavelength DIAL research has been conducted in Japan. Fukuchi and others [65, 66] investigated the use of multiple wavelengths to improve the accuracy of  $\text{SO}_2$  measurements to the 1-ppbv level at 300 m resolution, which they adopted as requirements for studying transport. No reported  $\text{SO}_2$  DIAL system had ever achieved this level of accuracy. They evaluated Dual DIAL using both three and four laser lines, and concluded that their desired accuracy should be achievable,



and that both aerosol effects and interference from ozone could be minimized by a proper choice of wavelengths.

Using a lidar system with two dual-wavelength tunable dye lasers, Fujii and others [67] actually achieved the predicted accuracy, measuring  $\text{SO}_2$  at 2400–3000 m altitude to 1 ppbv with a range resolution of 300 m. Their system, which they called *MDIAL* for Multiwavelength DIAL, transmitted 10–18 mJ per pulse in the 298–300 nm region, and the receiver was based on a 500-mm-diameter telescope. They typically averaged the lidar returns from 1500 laser shots at each wavelength. The lidar was also capable of simultaneous measurements of multiple species by conventional two-wavelength DIAL, and they used this feature to experimentally evaluate the effect of  $\text{O}_3$  on  $\text{SO}_2$  measurements.

In 2002, the same group of Japanese researchers [68] presented a further improvement in which they developed curve-fitting techniques for use with multi-wavelength lidar measurements. They used five wavelengths around an  $\text{SO}_2$  absorption peak, as shown in Fig. 7.10, to obtain both  $\text{SO}_2$  and  $\text{O}_3$  concentrations, and estimated that they could measure  $\text{SO}_2$  concentrations with errors less than 0.5 ppbv at altitudes of about 1 km, with 300 m resolution. The researchers also compared the curve

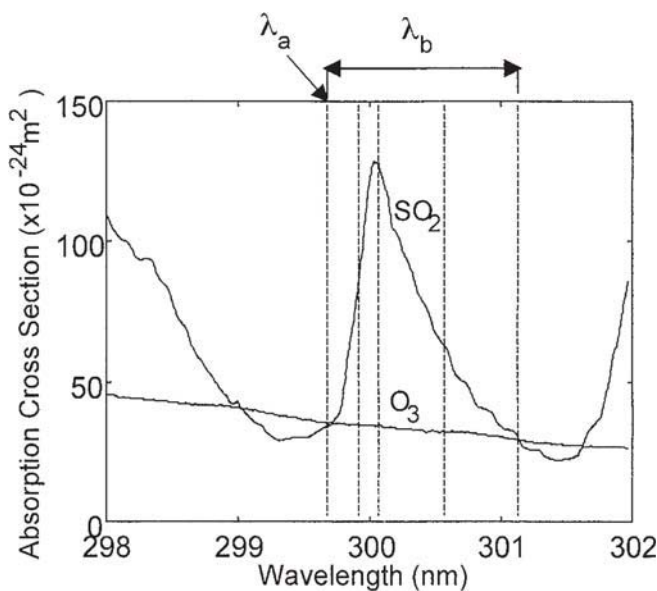


**Fig. 7.10.** Absorption cross sections for  $\text{SO}_2$  and  $\text{O}_3$  and the wavelengths used in the five-wavelength curve-fitting method for  $\text{SO}_2$  [66].

fitting technique with Dual DIAL, and improved their choice of Dual DIAL wavelengths. The disadvantage of the curve fitting technique was a long measurement time (25 min for five wavelength pairs).

## 7.5 Outlook and Conclusions

Any lidar technique is dependent on the availability of suitable lasers, but for DIAL, the requirements are especially inflexible because the required laser characteristics are determined by the spectra of the molecules to be measured. The biggest problem has historically been to develop reliable lasers with outputs at appropriate wavelengths. Fortunately, steady progress is being made with tunable laser sources, especially OPOs. Tunable lasers enable researchers to optimize the wavelength choices and in that sense they are better than fixed-frequency wavelength shifters such as Raman cells. In addition, OPOs hold the promise of compact, all-solid-state systems that can, in principle, be developed for any wavelength from the UV to the far IR.

Continued progress can be expected in the area of multiwavelength DIAL; this area of research has already produced an order of magnitude improvement in DIAL sensitivity. Ultimately, one might expect that instruments and algorithms will be developed to obtain range profiles of multiple gasses simultaneously, along with aerosol characteristics.

Hybrid techniques, such as the Doppler DIAL measurements by NOAA described earlier, have begun to emerge and this trend may continue. For example, the micropulse lidar concept [69] could be combined with DIAL to develop an eye-safe lidar for NO<sub>2</sub> operating in the visible region around 448 nm. An effort to develop a DIAL micropulse lidar for water vapor has already been reported [70].

Remote monitoring of localized pollution sources such as plumes was demonstrated very early in the history of DIAL, and lidar techniques were later demonstrated for measuring pollutant fluxes and emission rates from wide-area sources such as industrial plants, particularly by researchers in Sweden and at NOAA. Further applications to large sources such as agricultural facilities and landfills are likely in the future.

DIAL systems for ozone and industrial pollution will no doubt continue to gain acceptance as costs become lower, reliability gets better, and algorithms to provide reduced data in real time are implemented in software. Another important issue discussed by Weitkamp and others [71] is the development of guidelines. The German Commission

on Air Pollution Prevention, KRdL, published guidelines for DIAL in 1999 [72]. The guidelines provide a foundation for lidar quality assurance and they are intended to help prospective users in several different ways. Guidelines and standards will undoubtedly play a key role in gaining acceptance for widespread applications of DIAL systems for air quality and emission monitoring.

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## Differential-Absorption Lidar for Water Vapor and Temperature Profiling

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### 8.1 Introduction

The importance of water vapor in the atmosphere can hardly be overestimated. Water vapor is the most important greenhouse gas, much more effective than CO<sub>2</sub>, it governs the atmospheric water cycle which is the basis for life on earth, and it is a key component in atmospheric chemistry. The frequent occurrence of phase transitions from vapor to liquid water or ice crystals further enhances the importance of atmospheric humidity. Cloud formation and the various forms of precipitation certainly belong to the most important weather phenomena. The strong temperature dependence of the saturation vapor pressure in combination with vertical transport processes causes a large variability of the atmospheric humidity which exists on practically all scales from turbulence to global distribution.

In view of its importance the observation capabilities for atmospheric water vapor are clearly insufficient, both for the operational global observation system and for detailed process studies. Most routine observations are still made using *in situ* sensors on radiosondes. Apart from the problems caused by the sensor properties it is also the sampling strategy, typically only two instantaneous measurements per day for a relatively small number of stations worldwide, which does not permit a characterization of the water vapor distribution that comes even close to the requirements. Retrievals from spaceborne passive sensors can provide

some information, but vertical resolution in particular is insufficient in view of the frequent occurrence of strong vertical gradients.

For process studies the vertical structure of the atmosphere is of great importance. Observational possibilities for water vapor profiles that provide the necessary high resolution and accuracy are very limited. *In situ* measurements are possible from aircraft or helicopters, free flying or tethered balloons, and kites, but all of these have serious limitations especially for vertical profiling. Therefore remote sensing either from the ground or from aircraft is a highly appreciated solution of the observational problem, provided that good accuracy and resolution is attained.

Two lidar techniques can provide the required information on the water vapor vertical distribution with the necessary vertical and temporal resolution: Raman lidar and differential absorption lidar (DIAL). Raman lidar is treated in detail in Chapter 9 of this book. DIAL methodology and selected experimental results are presented here to provide an overview over the principal strengths and weaknesses of the method as well as its potential for applications in atmospheric research.

## 8.2 Methodology

The application of the DIAL technique as described in Chapter 7 to water vapor or temperature measurements does not pose any new fundamental problem, but there are some details that need to be considered carefully. This comes about mainly for two reasons: the accuracy requirements for water vapor and temperature retrievals are quite high, and the use of very narrow absorption lines of the rotational–vibrational spectrum in the near infrared makes the inversions prone to errors resulting even from small changes in the transmitted spectra or absorption spectra.

The primary result of the DIAL technique is the differential absorption coefficient, which is the product of the molecular differential absorption cross section and the molecule number density of the gas under study. From a measurement of the differential absorption coefficient, the density can be deduced if the differential absorption cross section is known, e.g., in water vapor profiling with DIAL. If the mixing ratio of a gas is known, e.g., for oxygen, the differential absorption cross section can be determined. Selecting an absorption line with a strong temperature dependence then allows the temperature profile to be obtained.

The common basis for water vapor and temperature profiling with DIAL is the use of isolated narrow absorption lines of the vibration-rotation spectrum of the water vapor or oxygen molecule, respectively. Therefore a brief summary of the spectroscopy of these gases is given below, mainly to quantify the pressure, temperature, and wavelength dependence of the absorption cross section. This helps assess the performance requirements for the main lidar components, the laser system and data acquisition.

### 8.2.1 Spectroscopy

The absorption coefficient  $\alpha_g$  of a gas depends on the molecule number density  $\varrho_n$  of the gas under study, the temperature, the partial pressures  $p_i$  of the components of the gas mixture, and the details of the transitions that contribute to absorption at the specified wavenumber. A general expression for the absorption  $\alpha_g$  at wavenumber  $\nu$  of a single absorption line centered at wavenumber  $\nu_0$  is given by

$$\alpha_g(\nu) = \varrho_n S(T, \epsilon) \Lambda(\nu - \nu_0, p_i, T) \quad (8.1)$$

where  $S(T, \epsilon)$  is the line strength of the transition at temperature  $T$  and initial-state energy  $\epsilon$ , and  $\Lambda(\nu - \nu_0, p_i, T)$  is the line shape function for wavenumber  $\nu$ , the partial pressures  $p_i$  of the components of the gas mixture, and temperature  $T$ . For water vapor and oxygen, the gases under consideration here, the dependencies are given as:

$$S(T, \epsilon) = S_0 \left( \frac{T_0}{T} \right)^l \exp \left[ -\frac{\epsilon}{k_B} \left( \frac{1}{T} - \frac{1}{T_0} \right) \right] \quad (8.2)$$

and

$$\Lambda(\nu - \nu_0, p, T) \approx \Lambda_V(\nu - \nu_0, p, T) = f' \mathcal{R}w(\xi + ia) \quad (8.3)$$

where  $S_0$  is the absorption line strength under standard conditions,  $k_B$  is Boltzmann's constant, and  $l$  is a constant that depends on the molecule ( $l = 1$  for  $O_2$  and  $l = 3/2$  for  $H_2O$ ).  $\Lambda_V$  is the Voigt absorption line function, which is a sufficiently good approximation to the actual line shape. The parameters for the Voigt function are given by  $f' = b_d^{-1} \sqrt{\ln 2 / \pi}$ ,  $a = b_c \cdot b_d^{-1} \cdot \sqrt{\ln 2}$ , and  $\xi = (\nu - \nu_0) \cdot b_d^{-1} \cdot \sqrt{\ln 2}$ , where  $b_c$  and  $b_d$  are the halfwidths (HWHM) for collision and Doppler broadening, respectively.  $\mathcal{R}w$  denotes the real part of the complex error function and  $i = \sqrt{-1}$ .



Two effects must be considered in the pressure dependence of the absorption cross section: collision broadening of the absorption line and shift of the line center with pressure. Pressure broadening is described by the collision broadening coefficient  $b_c$  in the Voigt function, it is different for each component of the gas mixture. Usually a single coefficient is given for air broadening which accounts for pressure broadening by nitrogen and oxygen. This is sufficient for a large altitude range where the mixing ratio remains constant. For tropospheric measurements water vapor pressure must also be considered, contributions from all other gases are negligible. It should be noted here that the self-broadening coefficient of water vapor is about 5 times higher than air broadening, so it is important even though its partial pressure is much smaller than the total pressure.

The pressure and temperature dependence of collision broadening by a single component of the mixture with partial pressure  $p_i$  is

$$b_{c,i}(p_i, T) = b_{c,i}(p_0, T_0) \frac{p_i}{p_0} \left( \frac{T_0}{T} \right)^{\eta_{c,i}} \quad (8.4)$$

where  $b_{c,i}(p_0, T_0)$  is the collision broadening coefficient at standard conditions. The effective collision-broadened width in a gas mixture is

$$b_c = \sqrt{\sum_i b_{c,i}^2(p_i)}. \quad (8.5)$$

The pressure shift of line center frequency with air pressure is described by a linear relation, the shift coefficient is temperature-dependent:

$$\nu_0(p_{\text{air}}, T) - \nu_0(p = 0, T_0) = d p_{\text{air}} \frac{p_{\text{air}}}{p_0} \left( \frac{T_0}{T} \right)^{\eta_{dp}} \quad (8.6)$$

The pressure shift induced by water vapor is different from that caused by air pressure, but is negligibly small under all atmospheric conditions.

The temperature dependence of the absorption cross section is of particular interest: for water vapor retrievals this dependence should be small to avoid errors due to insufficient knowledge of the temperature profile, and for temperature profiling this dependence should be as large as possible to increase the sensitivity of the method. Combining Eqs. (8.1)–(8.3) we derive

$$\frac{d\alpha}{\alpha} = \frac{dT}{T} \left[ \frac{\epsilon}{k_B T} - l - \frac{3}{2} + \Xi(\Lambda) \right]. \quad (8.7)$$

Here  $\Xi$  is a value that depends on the actual line shape and assumes values between 0 in the limit of pure Doppler broadening and 1 for pure collision broadening. It follows immediately that high temperature sensitivity is obtained for lines with a high initial-state energy  $\epsilon$ . Low temperature dependence for water vapor measurements is achieved for  $\epsilon/k_B T = 2$  when collision broadening prevails, and  $\epsilon/k_B T = 3$  when Doppler broadening is dominant.

For temperature profiling a tradeoff has to be made between line strength and temperature dependence, since with increasing initial state energy the lines become weaker, so that the measurable differential absorption becomes smaller. Lines that are useful for atmospheric measurements have temperature sensitivities of about  $1.4\% \text{ K}^{-1}$  (e.g., line  $^P P_{27,27}$ ) to  $2.4\% \text{ K}^{-1}$  (e.g.,  $^P P_{31,31}$ ). Obviously very accurate measurements of the differential absorption coefficient are required for an accuracy of better than 1 K in temperature retrieval. This can only be achieved if all details of the spectral distribution that affect the effective absorption coefficient are considered very carefully.

To be complete we note that small deviations from the Voigt line shape have been observed [1]. According to [2] the effect of these deviations on temperature retrievals is not very large, and it is even smaller for water vapor. Modified line shape functions have been developed which may be used if necessary.

In summary, the following parameters are needed for the calculation of the absorption cross section at wavenumber  $\nu$  for  $\text{H}_2\text{O}$  or  $\text{O}_2$  in an isolated absorption line at air pressure  $p_{\text{air}}$ , water vapor partial pressure  $p_{\text{H}_2\text{O}}$ , and temperature  $T$ :

|                               |                                                                                  |
|-------------------------------|----------------------------------------------------------------------------------|
| $\nu_0$                       | absorption line center under standard conditions                                 |
| $S_0$                         | absorption line strength under standard conditions                               |
| $l$                           | temperature exponent of the absorption line strength                             |
| $\epsilon$                    | initial-state energy of the transition                                           |
| $b_{\text{c,air},0}$          | collision broadening coefficient for air at standard conditions                  |
| $b_{\text{c,H}_2\text{O},0}$  | collision broadening coefficient for $\text{H}_2\text{O}$ at standard conditions |
| $\eta_{\text{c,air}}$         | temperature exponent of collision broadening for air                             |
| $\eta_{\text{c,H}_2\text{O}}$ | temperature exponent of collision broadening for water vapor                     |
| $dp_{\text{air}}$             | pressure shift coefficient for air                                               |
| $\eta_{dp}$                   | temperature coefficient of the pressure shift.                                   |

These parameters have been measured with high precision for a number of suitable lines, notably [1, 3–5]. Standard spectroscopy databases, e.g., HITRAN [6] or ESA [7], do not include all of these parameters, so the missing ones remain to be determined whenever a new absorption line is to be used for high precision DIAL measurements.

We note that the line strengths of water vapor are still under discussion. Significant changes for the HITRAN database have been introduced, and there are large differences between HITRAN and ESA, up to 30%. Since the sources for these differences have not yet been identified, a major uncertainty remains, although it is much smaller for the lines that are in use for DIAL work, because for these high-resolution studies based on tunable laser spectroscopy have been made as cited above, which presently appear to be the most reliable sources for the line parameters considered here.

Water vapor absorption lines are present in many regions of the infrared spectrum. For DIAL work the most suitable wavelengths are around 730, 820, and 930 nm, where interference with other gases is minimal, suitable laser sources and sensitive detectors are available, and a wide range of line strengths is covered. Figure 8.1 shows the absorption coefficient of water vapor in the 700 to 1200 nm region, for standard pressure and temperature and 80% relative humidity.

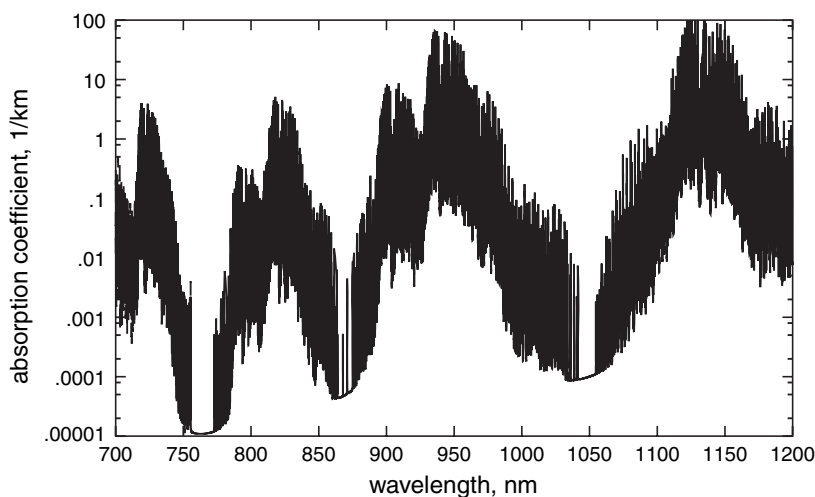


**Fig. 8.1.** Absorption coefficient of water vapor from 700 to 1200 nm. Standard pressure and temperature, 80% humidity.

### 8.2.2 Detailed DIAL Methodology

The use of the DIAL technique for narrow absorption lines requires detailed consideration of the spectral shapes of the transmitted and the backscattered radiation. While modern laser technologies can be used to generate extremely narrow lines with high spectral purity, the backscatter originating from molecular scattering always shows considerable Doppler broadening. Typical  $\text{H}_2\text{O}$  and  $\text{O}_2$  linewidths are plotted in Fig. 8.2 as a function of altitude in a standard atmosphere and compared with the widths of the Rayleigh line. It is obvious that the spectral shape of the Rayleigh-scattered light needs to be considered explicitly. For a full description of the problem the reader is referred to [8], a brief summary of the main results is presented here.

### The General Lidar Equation

The monochromatic form of the lidar equation reads [9]

$$P(v, R) = E_L \frac{c}{2} \frac{A}{R^2} \eta(v, R) \cdot \beta(v, R) \cdot e^{-2 \int_0^R \alpha(v, r) dr} \quad (8.8)$$

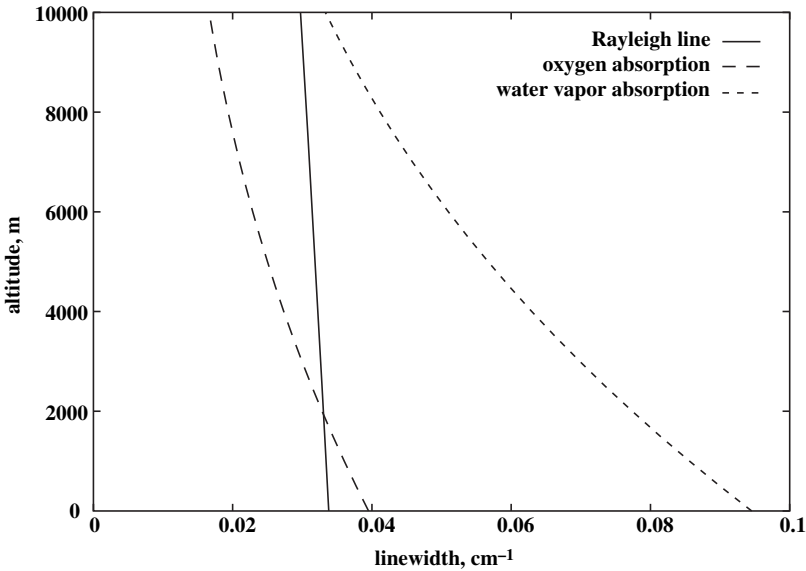


**Fig. 8.2.** Widths (HWHM) of the Rayleigh scattered line, the oxygen absorption line  $P_{27,27}$  at  $\nu_0 = 13010.81 \text{ cm}^{-1}$ , and the  $\text{H}_2\text{O}$  absorption line at  $\nu_0 = 13718.58 \text{ cm}^{-1}$ .

where  $P(\nu, R)$  is the signal power received from distance  $R$ ,  $\nu$  the wavenumber of the transmitted light,  $R$  the distance of the scattering volume from the transmitter/receiver,  $E_L$  the transmitted pulse energy,  $A$  the active area of the receiver telescope,  $\eta(\nu, R)$  the total system efficiency,  $\beta(\nu, R)$  the total backscatter coefficient at distance  $R$ , and  $\alpha(\nu, R)$  the total atmospheric extinction coefficient including gaseous absorption and molecular as well as particle scattering. We note explicitly that Eq. (8.8) is derived assuming instantaneous, incoherent, elastic single scattering. For specific applications the validity of these assumptions should be verified.

For a transmitter with an arbitrary spectral distribution  $l_t(\nu)$ , assumed to be nonzero in the interval  $\nu \in \Delta\nu$ , and normalized to  $\int_{\Delta\nu} l_t(\nu) d\nu = 1$ , Eq. (8.8) is integrated over the spectral distribution [10]:

$$P(R) = \frac{P_0}{R^2} \int_{\Delta\nu} l_t(\nu) \cdot \eta(\nu, R) \cdot \beta(\nu, R) \cdot \tau(\nu, R)^2 d\nu \quad (8.9)$$

where we make use of a lumped system constant and the atmospheric transmittance:

$$P_0 = E_L \cdot \frac{c \cdot A}{2} \quad \text{and} \quad \tau(\nu, R) = e^{-\int_0^R \alpha(\nu, r) dr} \quad (8.10)$$

Generally the spectral distribution may change during the scattering process, e.g., by inelastic scattering or by Doppler broadening of the Rayleigh backscatter. This can be treated in analogy to the treatment of the nonvanishing laser bandwidth by introducing a normalized spectral distribution after scattering,  $l_s(\nu - \nu', R)$ , where monochromatic excitation at  $\nu$  is assumed. Then the most general lidar equation (still assuming the case of incoherent, instantaneous, single scattering only) reads

$$P(R) = \frac{P_0}{R^2} \int_{\Delta\nu} \int_{\Delta\nu'} l_t(\nu) \cdot \eta(\nu', R) \cdot \tau(\nu, R) \cdot \beta(\nu, R) \\ \times l_s(\nu - \nu', R) \cdot \tau(\nu', R) d\nu' d\nu. \quad (8.11)$$

This equation is very general and is capable of handling complex laser emission as well as the full range of scattering processes with only the restrictions mentioned above. While Eq. (8.11) appears formally simple, the double integration can cause substantial problems, in particular when the equation is used for the inversion of measured lidar signals rather than for forward calculations.

For the applications considered here, i.e., humidity and temperature profiling, a few simplifications can be made. For these purposes laser sources with a very narrow transmitted spectrum should be used in any case. Then we may assume  $\eta(\nu)$  and  $\beta(\nu)$  to be constant for all  $\nu$  in the transmitted spectrum. Of the total extinction coefficient, which is the sum of extinction due to molecular scattering  $\alpha_m$ , particle scattering  $\alpha_p$ , and gaseous absorption  $\alpha_g$ , only gaseous absorption shows rapid spectral variations.

The spectral distribution function generally is a function of range  $R$  and differs for upward and downward propagation. For each spectral distribution an effective gaseous absorption coefficient can be defined as

$$\alpha_{\text{eff}}(R) = \int_{\Delta\nu} \alpha(\nu, R) l^*(\nu) d\nu. \quad (8.12)$$

Introducing the effective absorption coefficients for upward and downward propagation,  $\alpha_{u,\text{eff}}$  and  $\alpha_{d,\text{eff}}$ , which are generally different because of potentially different spectral distributions, and the correction term  $G$  which accounts for changes in the transmission of the back-scattered light on its way down from distance  $R$  due to a change in the spectral distribution, we can derive a lidar equation in differential form with direct physical interpretation:

$$\begin{aligned} \frac{d}{dR} \ln(P \cdot R^2) &= \frac{d}{dR} \ln \eta + \frac{d}{dR} \ln \beta - 2\alpha_p - 2\alpha_m \\ &\quad - \alpha_{u,\text{eff}} - \alpha_{d,\text{eff}} + G. \end{aligned} \quad (8.13)$$

The main difference compared to the standard lidar equation is the separation into upward and downward propagation, the introduction of effective extinction coefficients, and the correction term  $G$ .

## DIAL

The DIAL equation is obtained by combining the lidar equations for the two wavelengths used. Let us denote them by the index on for the wavelength at the center of an absorption line, called online, and the index off for the offline wavelength away from the line center. Let us further assume that we have chosen the offline wavelength sufficiently far from any other absorption line, but close enough to the online wavelength that the aerosol properties, backscatter and extinction, can be assumed the same, and in a region with slowly variable absorption coefficient such

that the details of the spectral distribution need not be considered. Under these conditions the resulting DIAL equation is relatively simple:

$$\frac{d}{dR} \ln \frac{P_{\text{on}}(R)}{P_{\text{off}}(R)} = \alpha_{\text{u,eff,on}} + \alpha_{\text{d,eff,on}} - 2\alpha_{\text{off}} + G_{\text{on}}. \quad (8.14)$$

The term  $\frac{d}{dR} \ln \frac{\eta_{\text{on}}}{\eta_{\text{off}}}$ , which describes the difference in the detection system sensitivity for the on- and offline wavelengths, has been omitted because this is considered as too specific for each individual system. A word of warning, however, appears appropriate: the narrow-band filters often used in DIAL receivers may in fact have different and range-dependent transmission for the two wavelengths. This results from the angular dependence of the filter transmission and the range dependence of the angular distribution of the collimated beam; for a detailed treatment see [11]. Although this effect can be corrected for, it is certainly preferable to avoid the problem by proper system design.

Equation (8.14) is the basis for water vapor retrievals as well as for temperature profiling. With modern laser techniques it is possible to make the transmitted spectrum sufficiently narrow so that  $\alpha_{\text{u,eff,on}}$  is given directly by the product of the absorption cross section and the number density:

$$\alpha_{\text{u,eff,on}} = \sigma_{\text{on}} Q_n. \quad (8.15)$$

This is also true for  $\alpha_{\text{off}}$ , which should be small anyway. For the calculation of  $\alpha_{\text{d,eff,on}}$  it is necessary to know the spectrum of the backscattered radiation and the absorption line shape as a function of altitude. For temperature-independent lines, which should be chosen for water vapor retrievals, this is straightforward using standard information for the estimation of the temperature and pressure profiles. The full treatment of  $G_{\text{on}}$  is beyond the scope of this chapter; for more details, the reader is referred to [8]. It may suffice to note that for its calculation the change in the backscatter spectrum must be known, which requires information about the scattering ratio profile.  $G_{\text{on}}$  is significant only in regions of steep gradients in aerosol backscatter, and is largest when molecular and particle scattering have about the same magnitude.

For temperature profiling the calculation of  $\alpha_{\text{d,eff,on}}$  is more complex because temperature affects both absorption line shape and line strength. However, an iterative solution with a standard starting profile converges very rapidly. Because of the higher accuracy requirements the problem of spectral broadening by molecular scattering is much more severe

for temperature retrievals. This is even more serious because regions of special interest such as layers of temperature inversion are typically associated with strong gradients in particle backscatter. Since this is a crucial issue for the applicability of temperature profiling with DIAL, it will be treated in more detail in Section 8.5.

When Eq. (8.14) is used to derive the water vapor density from measurements of  $P_{\text{on}}(R)$  and  $P_{\text{off}}(R)$ , it is necessary to know the parameters determining the absorption cross section, as listed in Section 8.2, and the spectral distribution of the transmitted and the backscattered light. If the transmitted spectrum is not much narrower than the absorption line under consideration, it must be specified very carefully.

A common problem for lasers with a broad tuning range is spectral impurity caused by amplified spontaneous emission, a broadband emission with typically very low spectral density which is hard to measure directly with standard spectroscopic techniques. We define spectral impurity as that fraction  $P_b$  of the total transmitted energy that is outside a well-defined laser line. Since this portion of the transmitted light will pass the atmosphere with practically no absorption, its contribution to the backscatter for the online wavelength will increase with optical tickness  $\tau_0$ . The relative error in the retrieved absorption coefficient is then

$$\frac{\Delta\alpha}{\alpha} = \frac{P_b}{P_b + (1 - P_b)e^{-\tau_0}}. \quad (8.16)$$

Only very small levels of spectral impurity can be tolerated when high accuracy is required. Another crucial point is that the laser must be tuned very precisely to the center of the absorption line. For small values of detuning the relative error in the effective absorption coefficient is given by

$$\frac{\Delta\alpha}{\alpha} = 1 - \frac{b_{\text{eff}}^2}{b_{\text{eff}}^2 + \Delta\nu^2}. \quad (8.17)$$

The requirements regarding laser properties for use with water vapor or temperature profiling are summarized in Table 8.1, as far as they can be derived from the spectroscopy of the absorption lines that are used. Specifications are such that individual errors remain <3% for water vapor and <0.6 K for temperature for worst-case conditions throughout the troposphere.



**Table 8.1.** Required laser performance for water vapor and temperature retrievals with individual errors of  $<3\%$  for water vapor and  $<0.6$  K for temperature.

| Parameter                                         | Requirement      |           |
|---------------------------------------------------|------------------|-----------|
|                                                   | H <sub>2</sub> O | T         |
| Laser linewidth, cm <sup>-1</sup>                 | $<0.013$         | $<0.0042$ |
| Frequency stability, $1\sigma$ , cm <sup>-1</sup> | $<0.007$         | $<0.0025$ |
| Spectral purity                                   | $>0.995$         | $>0.999$  |

### 8.3 Specific Solutions for Water Vapor DIAL Systems

Since the first application of the DIAL technique in 1966 [12], a number of systems for water vapor profiling have been described by several groups; for an overview over the developments before 1991 see [13]. The applicability of most systems before 1996 was severely limited by imperfections of the laser systems. Lack of wavelength stability and insufficient spectral purity were the most common problems. With the application of the injection seeding technique both problems could be overcome. In this technique a low-power stabilized cw laser provides the necessary spectral properties and is used to seed a power oscillator that provides the required pulse energy. This scheme is used successfully in different variants: Chyba [14] used a diode laser as a well-controlled cw source to seed a laser-pumped linear titanium:sapphire (Ti:Sa) power oscillator. Wulfmeyer [15] employed a laser-pumped Ti:Sa master oscillator and a flashlamp-pumped Alexandrite ring laser as a power oscillator. Ehret [16] reported the properties of an optical parametric oscillator, based on a Nd:YAG-laser and KTP as a nonlinear crystal. While all these developments have shown good performance in short-term missions, there is still need for further development. Operation requires too much effort for adjustment and maintenance, and long-term unattended operation has not yet been demonstrated. However, with the availability of reliable and affordable pump lasers (either diode- or flashlamp-pumped), with simplified resonator designs, ultra-stable mechanical setups for the resonator and the coupling of the subsystems, and automated system control, it appears feasible to overcome these problems in the next few years.

The second subsystem of a water vapor DIAL which has to meet very demanding specifications is the data acquisition chain from the detector to the analog-to-digital converter. No signal distortions  $\gtrsim 0.3\%$  can be tolerated, and a large dynamic range is required. The latter is different

for airborne or spaceborne systems looking downward and for ground-based systems looking upward. Figure 8.3 shows simulated lidar signals for a standard atmosphere [17] assuming constant relative humidity of 80% for all heights. For the aerosol backscatter an approximation to the average summer profile over Hamburg [18] is used, and an absorption line is chosen such that the optical depth of the troposphere due to water vapor absorption is 1. Plotted are online signals for a spaceborne lidar at 450 km altitude, a downward-looking airborne system at 12 km altitude, and a ground-based system looking upward. All signals are calculated for a nominal transmitted energy of 0.1 J, a telescope diameter of 0.5 m, and an overall system efficiency of 0.22. Nighttime conditions with no background light are assumed just to show the effect of the viewing geometry rather than to provide a complete performance simulation. The dynamic range the ground-based instrument must cover is enormous: six orders of magnitude. It is in particular the range below 2 or 3 km that causes substantial trouble, but that is also the region of special interest. For the downward-looking systems the situation is much easier. The airborne system needs to cover less than two orders of magnitude. The same holds for the spaceborne lidar, but the signal is very small for the parameters assumed here.

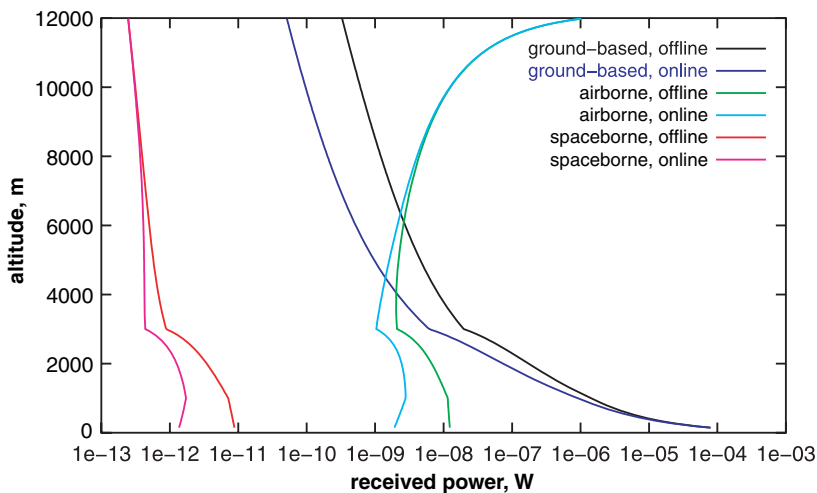


**Fig. 8.3.** Simulated lidar signals, online and offline, for a ground-based system, an airborne system flying at 12 km altitude, and a spaceborne system flying at 150 km altitude, all with the same technical specifications.

It must also be noted that the altitude range that can be covered by ground-based systems is limited to roughly 5 km, depending on the details of the meteorological situation. This is because most of the water vapor is located in the lowest 1–2 km, and the absolute humidity decreases by about three orders of magnitude between the top of the boundary layer and the tropopause. If a stronger line is chosen, the online signal becomes extremely weak because of water vapor absorption in the lower layers, and for a weak line extremely small differential absorption remains in the upper troposphere. Again the situation is much more favorable for downward-looking systems.

## **8.4 Applications of Water Vapor Profiling**

### **8.4.1 Assessment of Accuracy**

The key properties of water vapor profiling lidars for applications in atmospheric research and monitoring are accuracy, availability, range, and resolution. The areas of highest potential of water vapor profiling with DIAL are high-resolution studies of turbulent processes during daytime, ground-based monitoring of the lower troposphere during daytime, a variety of process studies using airborne systems, and, in the future, possibly spaceborne global monitoring. For turbulence studies high temporal and vertical resolution in combination with good relative accuracy are the essential features. For monitoring, absolute accuracy in combination with good availability and range are most important, and for airborne systems it depends on the specific application, but probably a combination of all properties is required.

The main properties for turbulence studies can be derived even without intercomparison to other systems using only sufficiently long periods of continuous lidar measurements. This is illustrated by an example described in [19]. The measurements were made in 1999 during an intercomparison with the Raman lidar at the Clouds and Radiation Testbed (CART) of the Atmospheric Radiation Measurement Program of the US Department of Energy (ARM) in Oklahoma. At that time this system (CARL) was one of the most advanced Raman lidar systems for routine humidity profiling [20]. To demonstrate the method of error assessment, Fig. 8.4 shows a variance spectrum of DIAL water vapor retrievals taken on October 9, 1999, 17:15 to 18:45 UT, at the ARM/CART site. It is a daytime measurement under clear-air convective conditions, altitude is

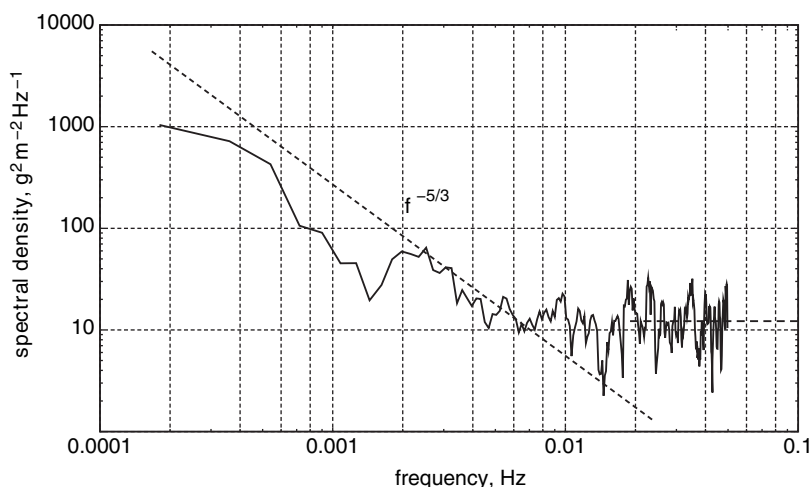


**Fig. 8.4.** Variance spectrum of the water vapor time series measured with DIAL during daytime. Roll-off according to  $f^{-5/3}$  (dotted line) and estimated noise level (dashed line) are indicated.

580 m above ground, temporal resolution 10 s, vertical resolution 75 m. The spectrum shows maximum values at low frequencies and a marked decrease toward higher frequencies. This decrease is proportional to  $f^{-5/3}$  over a substantial part of the spectrum as expected for the inertial subrange. The spectrum of random errors in the measurements should be white, i.e., constant over the whole frequency range. This is clearly visible in Fig. 8.4 for frequencies beyond 0.005 Hz. Even if the spectra do not show this typical pattern, an upper limit for the noise level can be estimated from the lowest statistically significant spectral density. Figure 8.5 shows two examples of noise levels determined in this way for MPI-DIAL and CARL, one during daytime and one during nighttime. For these cases resolution was chosen as 1 minute temporally and 90 m vertically. For the DIAL the estimated noise level is 3–7% during both day and night, increasing with height. During nighttime the Raman lidar shows about the same level in the near range and significantly less noise beyond 1 km. During daytime the Raman lidar performance is reduced considerably, noise level is at about 15%. Better performance of DIAL has been achieved on other occasions; nevertheless, this example demonstrates that high resolution in combination with good relative accuracy can be achieved during both day and night in the lower troposphere where turbulent processes are most important.

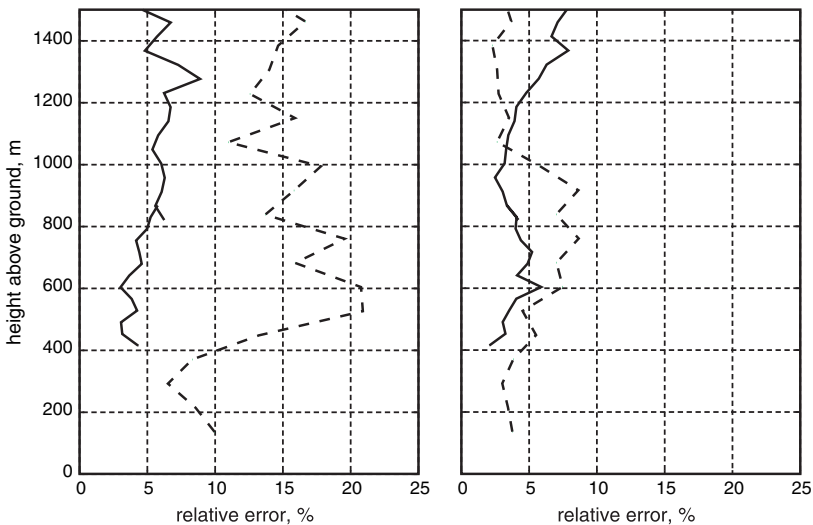


**Fig. 8.5.** Relative error of high resolution water vapor measurements for DIAL (solid) and Raman lidar (dashed) during daytime (left) and nighttime (right).

During the same campaign an attempt was made to compare the absolute accuracies of the MPI-DIAL, CARL, and radiosondes. We note here that Raman lidar does not provide absolute humidity measurements. The calibration of CARL relies on matching the total integrated water vapor to the results of a microwave radiometer operated at the same site. As an example Fig. 8.6 shows coincident profiles from a radiosounding, CARL, and MPI-DIAL measured during nighttime on October 10, 1999. Obviously there is very close agreement between all three systems up to about 8 km height, a height range in which water vapor density varies by two decades. Some deviations occur at layer boundaries, in particular in comparison of the two lidars with the radiosoundings. This illuminates one of the problems of intercomparisons, specifically with *in situ* sensors: mostly the sampled volume is not the same, and natural variability in the humidity distribution sets limits for this kind of intercomparison. The estimated errors for the two lidar soundings, presented in the right panel, clearly show that Raman lidars perform much better at night than in the daytime.

To come to a more generalized view of the differences between the systems, we compare the total water vapor content  $iwv$  integrated over a height range in which both systems operate reliably. Figure 8.7 shows this for the MPI-DIAL and CARL for 12-hour periods on 5 days, based on

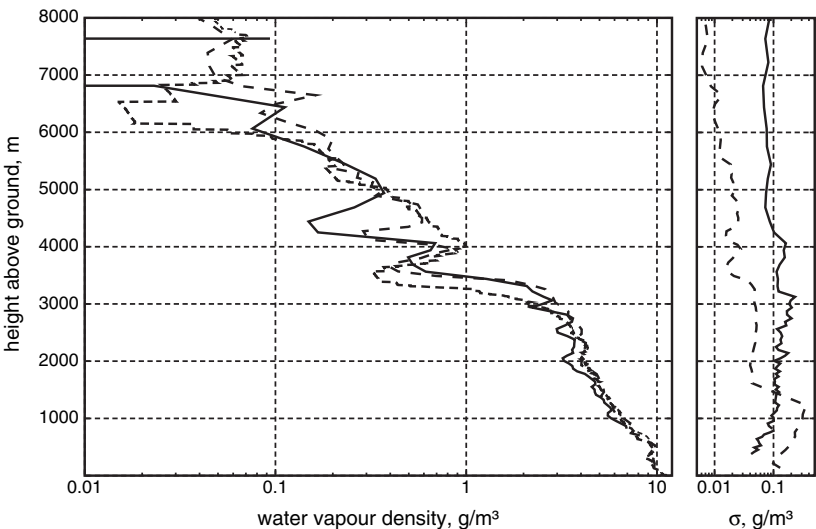


**Fig. 8.6.** Water vapour profiles (left) and estimated standard deviation (right) from radiosonde (dotted), Raman lidar (dashed) and DIAL (solid). October 10, 1999, 04:30 UT (nighttime).

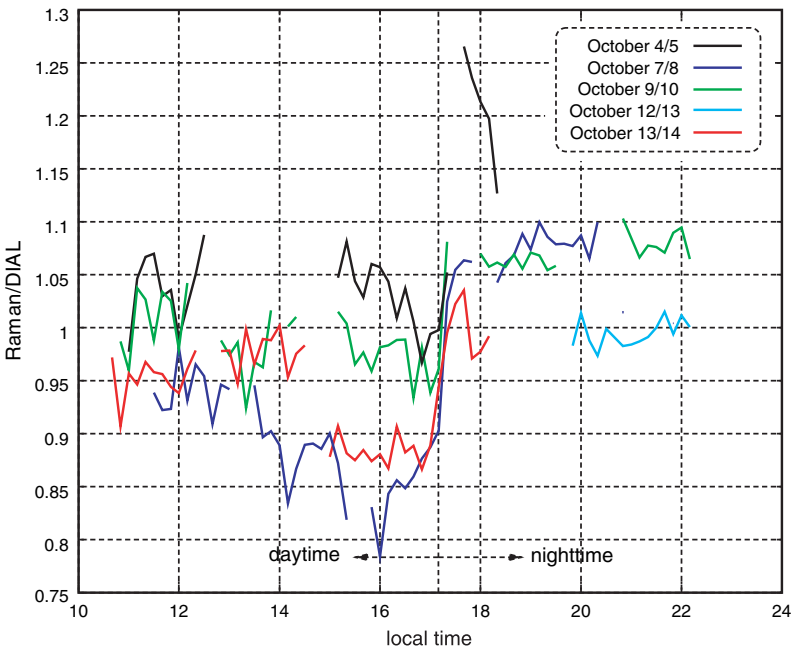


**Fig. 8.7.** Ratio of integrated water vapor measured by Raman lidar and DIAL for five 12-hour periods during the 1999 intercomparison campaign.

10-minute averages. During daytime the integration range extends from 1 to 3 km, during nighttime from 1 to 6 km. The ratio  $i_{wv}^{CARL}/i_{wv}^{DIAL}$  shows agreement to better than 10% with only few exceptions. On the average the  $i_{wv}$  is higher for the DIAL retrievals during daytime and lower during nighttime. There is considerable scatter mainly in the afternoon, and an abrupt change at 17 hours local time when CARL is switched from daytime to nighttime mode. During nighttime the difference appears to be either zero or around 7%. For the DIAL results occasionally a jump of a few percent occurs when the system is switched to a different absorption line.

The intercomparison results demonstrate that DIAL is favorable for high-resolution measurements in the lower troposphere during daytime, and that presently the relative uncertainty in absolute water vapor content is a few percent. Different calibration approaches and uncertainties in the absorption line parameters appear to contribute to the possible errors. These conclusions are also supported by other intercomparisons [21, 22].

#### 8.4.2 Turbulence Studies in the Atmospheric Boundary Layer

Studies of the turbulent boundary layer require daytime measurements of the humidity distribution with high accuracy and resolution. An example is presented here to demonstrate the capabilities of DIAL for this purpose [23].

Figure 8.8 shows the time–height distribution of the water vapor density measured on September 13, 1996, 07:00 to 08:15 UT. Measurements were made with the MPI-DIAL at the SE shore of the island of Gotland. The distribution shows mainly two different height regions, one with relatively high water vapor density around  $6.5 \text{ g/m}^3$  up to about 600 m height, and a much drier region above where the water vapor density is smaller than  $4.5 \text{ g/m}^3$ . The boundary between these two layers changes rapidly, updrafts of humid air are observed as well as downdrafts of dry air, both with varying dimensions. The mixing zone extends from about 300 to more than 700 m height. The pattern shows clearly that rather strong turbulence occurred during this time period, and that the strong wind of 12 m/s advected the eddy structures rapidly. It is the high-temporal resolution of the DIAL, 10 s in this case, that enables us to resolve these structures.

Figure 8.9 shows humidity variance spectra at selected heights for the case shown in Fig. 8.8. At lower heights, 270–510 m, the spectra show a weak maximum at about 0.001 Hz corresponding to a period of

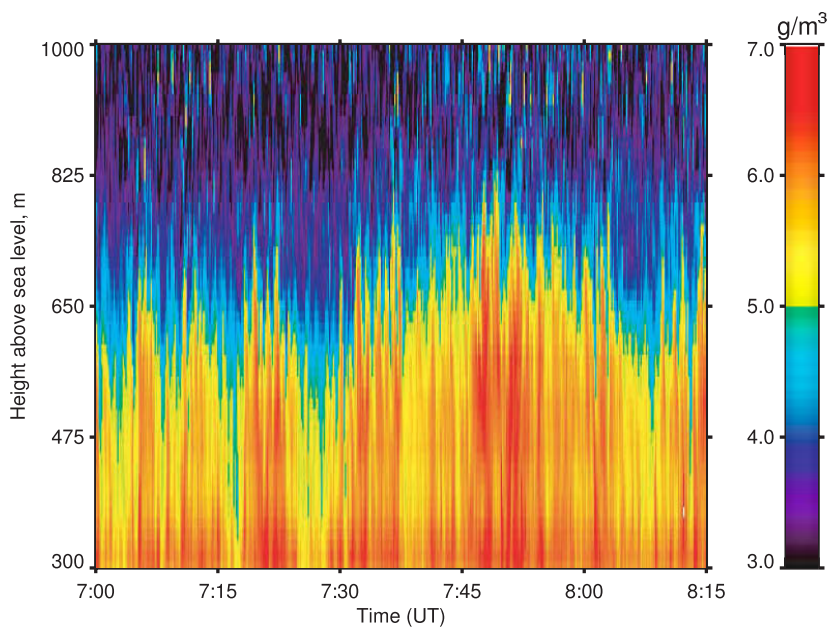


**Fig. 8.8.** Time-height cross section of the water vapor density. Temporal resolution is 10 s, vertical resolution is 60 m. Gotland, September 13, 1996. From [23].

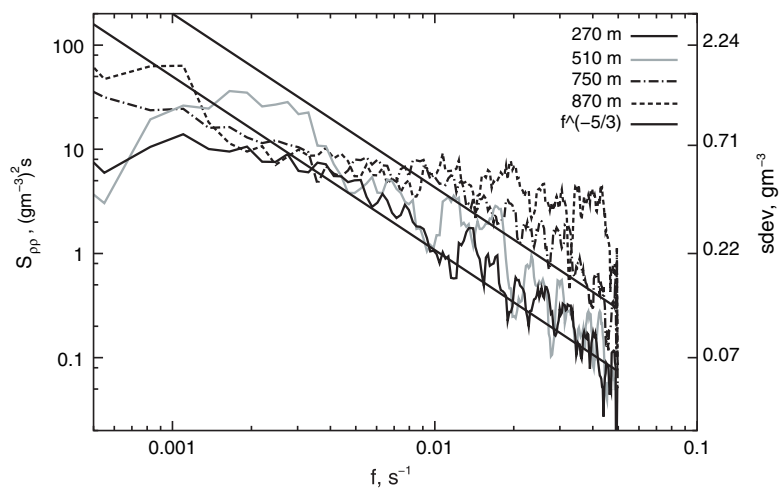


**Fig. 8.9.** Humidity variance spectra at selected heights. Gotland, September 13, 1996. From [23].



17 minutes, and then roll off approximately proportional to  $f^{-5/3}$  up to the Nyquist frequency of 0.05 Hz. This is as expected and consistent with the assumption that for frequencies  $>0.005$  Hz the inertial subrange is reached in which turbulence energy is mainly transported from large to small eddies. At the high-frequency end these spectra still show a rolloff proportional to  $f^{-5/3}$ , there is no indication of noise. Apparently noise is  $<0.07$  g/m<sup>3</sup> rms at these height levels. At an altitude of 750 m the variance spectral density is up to a factor of 4 larger, in particular at frequencies beyond 0.004 Hz, but the same rolloff with  $f^{-5/3}$  is observed, again with no indication of noise. At 870 m the variance is quite similar in the low-frequency range, but the decrease with frequency is much weaker. This can be explained by an increased noise contribution to the variance, about 0.4 g/m<sup>3</sup> rms. A strong increase in system noise is expected for height levels beyond the top of the boundary layer because of reduced aerosol backscatter.

It is interesting to inspect the probability distribution functions for water vapor density as shown in Fig. 8.10 at selected heights for the case under study. At lower heights the distribution exhibits a rather sharp peak at 6–6.5 g/m<sup>3</sup>. When the lower end of the entrainment zone is reached, e.g., at 510 m, the distribution shows additional broadening at lower humidity values. In the middle of the entrainment zone, at 630 m,

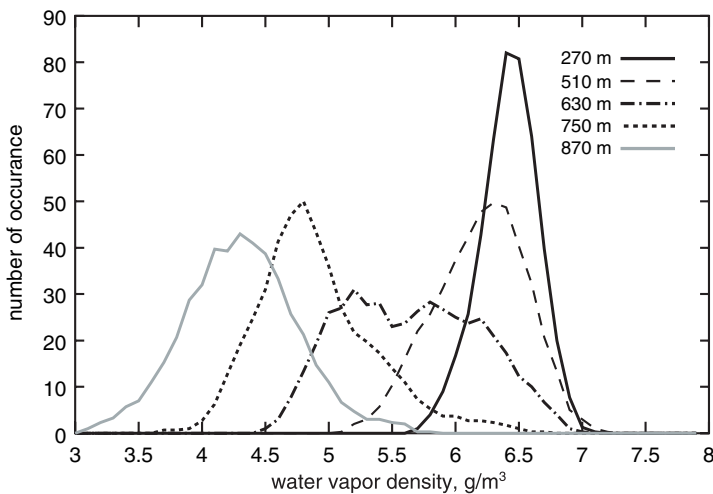


**Fig. 8.10.** Probability distribution functions for water vapor density at selected height levels. Gotland, September 13, 1996. From [23].

the distribution is characteristic for a mixture between a distribution centered at about  $6 \text{ g/m}^3$  and another one centered at about  $5.3 \text{ g/m}^3$ . At greater heights the distributions get more localized again at lower humidity values before they spread out because of noise contributions. This example clearly demonstrates that profiles of distribution functions are well suited to characterize the mixing processes in the boundary layer. DIAL measurements can provide this information.

The combination of high-resolution profiling for humidity and the vertical wind component allows one to determine profiles of the latent heat flux using the eddy-correlation technique [24]. In this technique, which is widely used for *in situ* flux measurements, the flux  $F_q$  is determined directly as the product of the water vapor density  $q$  and the vertical velocity  $w$ :

$$F_q = \overline{q \cdot w}. \quad (8.18)$$

By definition the product is the instantaneous local flux, but averaging in time and/or space is necessary to provide a value that is representative for a certain period and area. This averaging is indicated by the overbar in Eq. (8.18). This method of direct flux measurement has the advantage that it does not depend on any assumption about the turbulence structure. However, some care needs to be taken in the estimation of representativeness. Only if a sufficiently large number of eddies has passed over the system can  $F_q$  be assumed representative for a larger area, otherwise the result may be rather random with even its sign depending on the actually observed part of the eddies.

So far the method has been used only occasionally, mainly with a combination of DIAL for the density measurements of water vapor or ozone and a Radio Acoustic Sounding System (RASS) for the vertical wind measurements [24, 25]. Combination of DIAL with a Doppler lidar has also been reported [26], which promises much better match of the sampling volumes and much better height coverage. Attempts are presently made to explore the possibility of measuring both quantities with the same system, a Doppler lidar with DIAL capability [27, 28].

The use of the eddy correlation method with remote sensing instruments will find broader application only if the system complexity and the effort needed for combined system operation is reduced considerably. The attempt is worth being made because it is a unique way to measure flux profiles in the boundary layer over rather long periods (as compared to, e.g., aircraft measurements).

### 8.4.3 Airborne Water Vapor Profiling

The possibility of building water vapor DIAL systems that can be operated on board an aircraft has attracted atmospheric scientists from the beginning. In fact more laboratories were involved in the development of airborne DIAL than in ground-based systems [29–31]. The main advantages of airborne over ground-based systems certainly are the flexibility in choosing the geographical region for experiments, in particular regions which would otherwise be hard if not impossible to access, and to cover large areas or to follow an object of interest. It also allows us to look down on the lower troposphere while simultaneously looking up to the upper troposphere/lower stratosphere without being obstructed by the dense layer of water vapor near the ground. A series of experiments that exploited these possibilities to a large extent was organized by NASA in the frame of the Convection And Moisture EXperiment (CAMEX) to investigate the distribution of water vapor, aerosols, clouds, and precipitation around hurricanes. During these experiments an advanced, fully engineered, automated water vapor DIAL, the Laser Atmospheric Sensing Experiment (LASE) [29], was operated on board a DC-8 aircraft. It is based on a Ti:sapphire laser, which is injection seeded with a diode laser frequency-locked to a strong water vapor line in the 815-nm band. A special feature of the laser transmitter is that the seeder can be tuned electronically to any spectral position on the absorption line to choose the optimal absorption cross section for the scene to be investigated. Fast switching between different positions is possible, permitting the use of different absorption cross sections in a rapid sequence. This allows one to cover the wide range of several decades of water vapor density that is found in a single column, in particular because the system is both looking downward into the lower troposphere and upward into the upper troposphere/lower stratosphere.

Figure 8.11 shows a GOES-8 image of hurricane “Bonnie” close before landfall on August 26, 1998, with the flight track of the DC-8 carrying LASE superimposed. It is obvious that such an extended and fast-developing weather system can only be investigated with an airborne instrument in such a way that all important parts of the system are observed in about the same status of development.

As an example for the results that can be achieved Fig. 8.12 shows a cross section of the first flight leg, tangential to the SE part of the storm. In the aerosol distribution, not shown here, the narrow rain band that was traversed around 11:50 UT is clearly visible. Here cloud tops exceeded

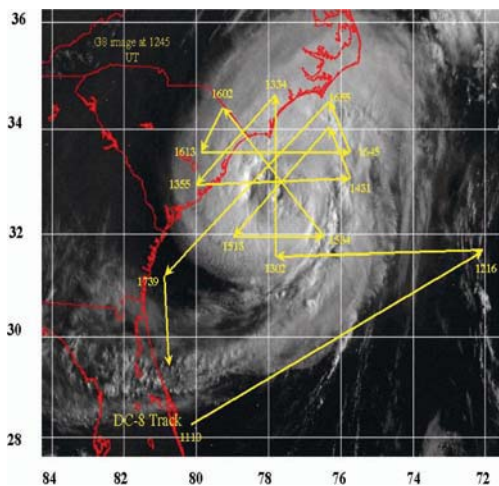


**Fig. 8.11.** GOES-8 image of hurricane “Bonnie” on August 26, 1998, with the flight track of the DC-8 carrying LASE superimposed.

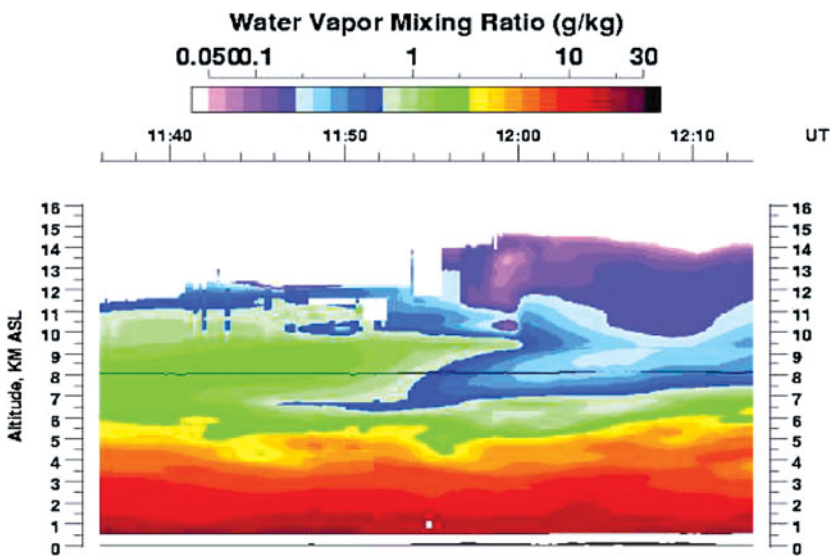


**Fig. 8.12.** Water vapor distribution on the flight leg from 11:28 to 12:13 UT. The horizontal black line at 8 km indicates the flight level. Horizontal resolution is 14 km (about 1 min), vertical resolution is 330 m for the lower part and 550 m for the middle and upper troposphere.

7 km altitude. The marine boundary layer in the inflow region of the hurricane is also well determined, it extends to about 1 km height. The specific humidity exceeds 15 g/kg in the boundary layer, and a region of moist air extends up to about 5 km, remarkably higher than the aerosol layer. In the upper troposphere the region to the NE of the rain band is clearly drier than the region to the SW of it, a fact that is also reflected in the enhanced cloud cover beyond 8 km in that sector. For further results and a detailed discussion the reader is referred to [32].

The example clearly shows that lidar measurements with airborne systems can make a unique contribution to studies of important weather phenomena that cannot be obtained by other methods. Airborne DIAL has been applied to studies of several other atmospheric phenomena that range from boundary layer processes to stratospheric intrusions. With the level of maturity that has been reached for the methodology and technology it is expected that these systems will be employed in a large variety of dedicated studies of atmospheric processes.

## 8.5 Temperature Profiling

The potential of the DIAL technique for temperature profiling was first introduced by Mason [33] and developed further by Schwemmer and Wilkerson [34], Korb and Weng [35, 36], and Mégie [10]. To the author's knowledge only one attempt to perform range-resolved temperature measurements with this technique has been reported so far [2].

The method is based on Eq. (8.1), which describes the dependence of the absorption coefficient on the number density of the absorber, the temperature-dependent strength of the absorption line, and the temperature- and pressure-dependent line shape. In most DIAL applications this is used to determine the density of the absorbing gas from the measured absorption coefficient and the known line strength and line shape, but if the absorber density is known it can also be used to determine the temperature-dependent line strength and from that the temperature itself. For this method oxygen is used as an absorber because it is known to have a constant mixing ratio in the atmosphere up to high altitudes, and because it has suitable absorption lines in an easily accessible part of the spectrum.

Although the method appears simple in principle, a more detailed look shows that several problems need to be addressed. Let us first rewrite

Eq. (8.1) for direct application to temperature profiling:

$$\alpha(\nu, p, T) = q_{\text{O}_2}(1 - q_{\text{H}_2\text{O}}) \frac{p}{k_{\text{B}}T} S(T, \epsilon) \Lambda(\nu - \nu_0, p_i, T), \quad (8.19)$$

where  $q_{\text{O}_2}$  and  $q_{\text{H}_2\text{O}}$  denote the mixing ratios of oxygen and water vapor, respectively. The first obvious complication is that the water vapor profile has to be known, too, which calls for a combination with a water vapor DIAL. Second, Eq. (8.19) represents a nonlinear relation between  $\alpha$  and  $T$  which cannot be solved for  $T$  analytically. However, there is a robust and fast converging iterative solution [36] which will not be given in detail here. It must also be noted that the atmospheric pressure profile needs to be known, which is generally calculated from the measured pressure at ground level and the temperature profile. Because the latter is initially unknown, again an iterative procedure is involved which also converges fast.

In the choice of a suitable absorption line, a trade-off must be made between high temperature sensitivity of the absorption cross section, which is largest for high initial-state energy, and a suitable magnitude of the absorption coefficient, which decreases with increasing initial-state energy. It turns out that suitable lines have a temperature sensitivity of the absorption cross section on the order of only 1–3%  $\text{K}^{-1}$ . This implicates that the absorption coefficient must be determined with less than 1% error to retrieve the temperature with the required accuracy of better than 1 K. This makes clear why temperature profiling using DIAL is extremely demanding systematically as well as technically. It also makes clear why this technique has so far not been used in practical applications.

It is beyond the scope of this chapter to discuss all possible systematic errors; for this, the reader is referred to [2]. There it is demonstrated theoretically as well as experimentally that the problem of the insufficiently known contribution of Doppler-broadened Rayleigh scattering to the total signal is the main source of uncertainty of the resulting temperature profile, provided that all other systematic and experimental errors have been reduced to the greatest possible extent. While in regions of dominating aerosol backscatter, specifically the well-mixed boundary layer, the observed errors were below 1 K, the observed temperature error exceeded 3 K at the top of the boundary layer where strong gradients in aerosol backscatter were observed. It is probably because of these difficulties, in combination with the availability of other measurement

methods, see e.g., Chapter 10 of this volume, that no further attempts to use the DIAL technique for temperature profiling have been reported.

## 8.6 Conclusions

The application of differential absorption lidar to narrow lines of the rotational-vibrational spectrum of water vapor or oxygen for humidity and temperature profiling is technically demanding with respect to the laser source and the data acquisition. Many details need to be considered carefully in system design and data evaluation. If that is done properly the technique is very powerful, in particular for water-vapor profiling. The main strengths of DIAL in ground-based applications are its excellent daytime performance for high-resolution studies in the boundary layer and high-accuracy routine observations in the lower half of the troposphere, as well as its independence from external calibrations. Suitability for airborne and probably also for spaceborne applications is definitely another very important feature of the method.

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## Raman Lidar

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### 9.1 Introduction

The Raman lidar technique makes use of the weak inelastic scattering of light by atmospheric molecules [1–8]. The excitation of a variety of rotational and vibrational molecular energy levels leads to several bands of Raman scattered radiation the frequency shifts of which are characteristic for the interacting molecule. Raman lidar systems have become widely used instruments in atmospheric research during the past decade [9–16]. The very robust technique makes low demands concerning spectral purity of the emitted laser light and frequency stabilization of the receiver. However, it suffers from the low cross sections of Raman scattering and thus from the comparably small signal-to-noise ratios of the measurements. For a long time, Raman lidar instruments were therefore mainly used at nighttime. Daytime applications increased with the development of high-power transmitters and narrow-bandwidth detection systems which allow a sufficient suppression of the daylight background [11, 17–19].

Raman measurements do not require specific laser wavelengths as it is the case in the differential-absorption lidar (DIAL) technique (see Chapters 7 and 8). Because of the wavelength dependence of the Raman scattering cross section which is proportional to  $\lambda^{-4}$ , with  $\lambda$  denoting the wavelength of the laser light, shorter emission wavelengths are to be preferred. Attenuation of the laser light by gaseous, especially ozone, absorption can be avoided if wavelengths  $\geq 320$  nm are chosen. However, the solar-blind region below 300 nm has also been used for Raman measurements to avoid daylight background [20–29]. The signal attenuation by ozone absorption limits the range of these measurements to a few

kilometers. Concerning measurement range, laser wavelengths between 320 and 550 nm are best suited for Raman applications. In the visible spectral region, the higher atmospheric transmission, i.e., the lower signal extinction by Rayleigh scattering which is also proportional to  $\lambda^{-4}$ , partly compensates for the lower Raman scattering cross sections.

The low Raman scattering cross sections require comparably high concentrations of the investigated atmospheric components. Accordingly, nitrogen, oxygen, and water vapor as main constituent gases in air are of prime interest. Whereas nitrogen and oxygen, the atmospheric concentrations of which are known in principle from temperature and pressure, serve as reference gases, the water-vapor concentration is a major unknown in atmospheric studies and therefore a principal subject of Raman lidar investigations. For that reason, this chapter will mainly focus on Raman observations of atmospheric moisture. Several attempts have been made to measure other gaseous constituents of the atmosphere. Carbon dioxide [9, 30] and atmospheric pollutants such as sulfur dioxide [30, 31] and methane [32, 33] have been investigated. However, the detection limits necessary to allow the application of the technique in routine atmospheric monitoring are hard to reach and the achievable measurement ranges are low. One of the topics which gained further interest in recent years is the detection of Raman scattering from liquid water in tropospheric clouds [34–36].

Several Raman lidar applications are based on the measurement of profiles of the reference gases nitrogen and oxygen. The temperature dependence of the rotational Raman bands of these gases permits one to determine atmospheric temperature profiles from the ground up to about 40 km height [5, 19, 37–41]. This technique is further explained in Chapter 10. The measurement of extinction and backscatter coefficients and thus of the extinction-to-backscatter ratio, or lidar ratio, of aerosols and optically thin clouds makes use of the fact that the Raman backscatter coefficient of the reference gas is known and the lidar equation can therefore be solved for the unknown particle extinction coefficient [42, 43]. The wide field of research based on this technique is discussed in Chapter 4. Two Raman nitrogen and/or oxygen signals, one of which is partly absorbed by ozone, are used to determine ozone concentrations using the so-called Raman DIAL technique [44–46].

Table 9.1 gives an overview of the Raman lidar techniques and their typical achievable measurement ranges under consideration of the latest technical developments. The use of the rotational Raman (RR) and vibration—rotation Raman bands (VRR) is discussed in more detail below.

**Table 9.1.** Overview of Raman lidar techniques. VRR – vibration–rotation Raman band, RR – rotational Raman band, SB – solar-blind spectral region

| Measured quantity                | Interacting molecule, Raman band used                       | Typical achievable measurement range              |
|----------------------------------|-------------------------------------------------------------|---------------------------------------------------|
| Water-vapor mixing ratio         | H <sub>2</sub> O (vapor), VRR<br>(+ reference gas, VRR)     | 0–12 km (night) [16, 47]<br>0–5 km (day) [11, 47] |
| Extinction coefficient           | N <sub>2</sub> or O <sub>2</sub> , VRR or RR                | 0–30 km (night) [48]                              |
| Backscatter coefficient          | (+ elastic signal for backscatter and lidar ratio)          | 0–10 km (day) [49]                                |
| Lidar ratio                      |                                                             |                                                   |
| Temperature                      | N <sub>2</sub> and/or O <sub>2</sub> , RR                   | 0–40 km (night) [19, 40]<br>0–12 km (day) [19]    |
| Ozone concentration (Raman DIAL) | N <sub>2</sub> and/or O <sub>2</sub> ,<br>VRR-VRR or RR-VRR | 3–20 km (night) [46]<br>0–3 km (day SB) [29]      |
| Other trace-gas concentrations   | Specific gas, VRR<br>(+ reference gas, VRR)                 | 0–1 km (day and night) [32]                       |
| Liquid water                     | H <sub>2</sub> O (liquid), VRR<br>(+ reference gas, VRR)    | 0–4 km (night) [34]                               |

In the following Section 9.2, the basic principles of Raman scattering are explained. Section 9.3 describes technical requirements and principal features of a Raman lidar system. The determination of water-vapor profiles and their errors is discussed in Section 9.4. Section 9.5 briefly introduces the Raman ozone DIAL technique and the measurement of liquid water. Finally, a few concluding remarks are given in Section 9.6.

## 9.2 Basic Principles of Raman Scattering

### 9.2.1 Frequency Shifts

The scattering of incident radiation by atmospheric molecules involves elastic and inelastic processes. We speak of elastic or Rayleigh scattering if the frequency of the scattered photon  $\tilde{\nu}_s$  is the same as the frequency of the incident photon  $\tilde{\nu}_i$ . In this case the molecule preserves its vibration–rotation energy level during the scattering process. Inelastic or Raman scattering processes lead to a change of the molecule’s quantum state, and the frequency of the scattered photon is shifted by an amount  $|\Delta\tilde{\nu}|$  [50–52]. If the molecule absorbs energy, i.e., a higher energy level is excited, the frequency of the scattered photon is decreased,  $\tilde{\nu}_s = \tilde{\nu}_i - |\Delta\tilde{\nu}|$ , the wavelength is red-shifted. We call this inelastic process Stokes Raman scattering. If the molecule transfers energy to the scattered photon by decreasing its energy level, the frequency of the scattered photon is

increased,  $\tilde{\nu}_s = \tilde{\nu}_1 + |\Delta\tilde{\nu}|$ , the wavelength is blue-shifted, and we speak of anti-Stokes Raman scattering. The shift

$$\Delta\tilde{\nu} = \tilde{\nu}_1 - \tilde{\nu}_s = \frac{\Delta E}{hc_0} \quad (9.1)$$

is characteristic for the scattering molecule. Here  $\Delta E$  is the energy difference between the molecular energy levels involved,  $h$  is Planck's constant, and  $c_0$  the speed of light in vacuum. (For the sake of consistency with the spectroscopic literature, we use the wavenumber  $\tilde{\nu} = 1/\lambda$  and the wavenumber shift  $\Delta\tilde{\nu}$  both with the unit  $\text{cm}^{-1}$  to describe the frequency and frequency shift of radiation. We follow, as far as possible, the recent book by D.A. Long [53] which we recommend for further reading.)

The calculation of molecular energy levels is straightforward for homonuclear diatomic molecules, in our case  $\text{N}_2$  and  $\text{O}_2$  [53–57]. For such molecules the approximation with the model of a freely rotating harmonic oscillator gives the energy of the vibrational levels:

$$E_{\text{vib},v} = hc_0\tilde{\nu}_{\text{vib}}(v + 1/2), \quad v = 0, 1, 2, \dots, \quad (9.2)$$

with the specific vibrational wavenumber or oscillator frequency of the molecule  $\tilde{\nu}_{\text{vib}}$  and the vibrational quantum number  $v$ .

For the rotational energy levels we get in a good approximation

$$E_{\text{rot},J,v} = hc_0[B_v J(J+1) - D_v J^2(J+1)^2], \quad J = 0, 1, 2, \dots \quad (9.3)$$

$J$  is the rotational quantum number, i.e., a series of rotational quantum levels belongs to each vibrational level.  $B_v$  is the specific rotational constant and  $D_v$  the centrifugal distortion or stretching constant of the molecule. The constants  $B_v$  and  $D_v$  depend on the actual vibrational state  $v$  of the molecule. The term with  $D_v$  considers the centrifugal stretching of the molecule's axis because of rotation. Its relative contribution to  $E_{\text{rot},J,v}$  is small and plays a role only for high  $J$  [57]. The molecular constants  $\tilde{\nu}_{\text{vib}}$ ,  $B_0$ ,  $B_1$ , and  $D_0$  needed to calculate Raman frequency shifts of  $\text{N}_2$  and  $\text{O}_2$  for Raman lidar applications are given in Table 9.2 [56–61].

A certain vibration–rotation energy level of the molecule is calculated from the sum of Eqs. (9.2) and (9.3). When applying these equations for the calculation of frequency shifts  $\Delta\tilde{\nu}$  after Eq. (9.1), we have to consider the selection rules for vibrational and rotational transitions, which are

$$\Delta v = 0, \pm 1 \text{ and } \Delta J = 0, \pm 2, \quad (9.4)$$

**Table 9.2.** Molecular constants for the calculation of Raman frequency shifts of nitrogen and oxygen. The values for  $\tilde{\nu}_{\text{vib}}$  consider an anharmonicity correction for the transition from the ground state to the first excited vibrational state (see Ref. [53], p. 125 and 182)

| Gas            | $\tilde{\nu}_{\text{vib}}$<br>$\text{cm}^{-1}$ | $B_0$<br>$\text{cm}^{-1}$ | $B_1$<br>$\text{cm}^{-1}$ | $D_0$<br>$\text{cm}^{-1}$  |
|----------------|------------------------------------------------|---------------------------|---------------------------|----------------------------|
| N <sub>2</sub> | 2330.7                                         | 1.98957 [59]              | 1.97219 [59]              | $5.76 \times 10^{-6}$ [59] |
| O <sub>2</sub> | 1556.4                                         | 1.43768 [58]              | 1.42188 [61]              | $4.85 \times 10^{-6}$ [58] |

with  $\Delta J = J' - J''$ . Here  $J'$  is the quantum number of the higher and  $J''$  the one of the lower energy level involved in the transition, independent of which of the levels is the initial or the final one. In contrast,  $\Delta v$  is the difference of the vibrational quantum numbers of the final and the initial vibrational states.

Figure 9.1 depicts schematically the transitions between different vibration–rotation energy levels of the N<sub>2</sub> molecule and the resulting Raman spectrum. The transitions with  $\Delta v = 0$  and  $\Delta J = 0$  correspond to Rayleigh scattering. Transitions with  $\Delta v = +1$  lead to the Stokes vibration–rotation band, and those with  $\Delta v = -1$  to the anti-Stokes vibration–rotation band. If the rotational quantum number does not change during the vibrational transition, i.e.,  $\Delta J = 0$ , the resulting Raman lines have very small frequency shifts between each other which are usually not resolved in lidar applications, and the group of lines is called the Q branch. Changes of  $\Delta J = +2$  and  $\Delta J = -2$  lead to the S and O branches, respectively. The rotational branches to the sides of the Rayleigh line are both of S type, since  $J' > J''$  if  $\Delta v = 0$ . We call these lines the Stokes and anti-Stokes (pure) rotational Raman lines. From Eqs. (9.3) and (9.4) and by neglecting the centrifugal stretching, we obtain that the rotational Raman lines are equidistant. The first line is shifted from  $\tilde{\nu}_1$  by  $6B_0$ , the next lines follow in distances of  $4B_0$ .

### 9.2.2 Cross Sections

The intensity of an observed Rayleigh or Raman line depends on the cross section of the corresponding vibration–rotation scattering process which is the product of the transition probability and the population of the initial energy level. For lidar applications we need the differential cross section  $d\sigma(\pi)/d\Omega$  for scattering at  $180^\circ$ , which we call the backscatter

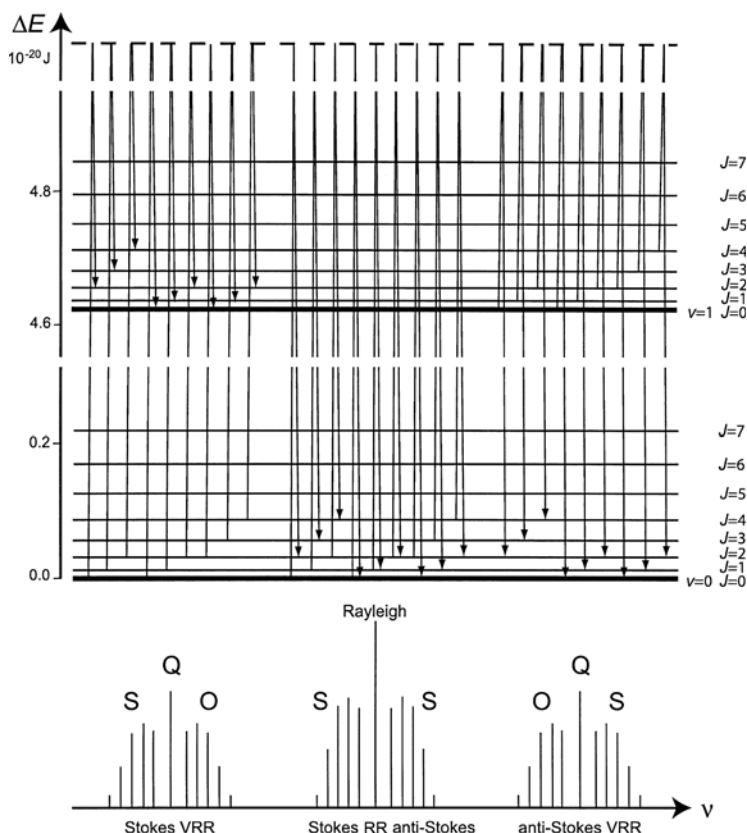


**Fig. 9.1.** Vibration-rotation energy levels of the  $N_2$  molecule, Raman transitions, and resulting spectrum.

cross section. Cross sections are calculated from Placzek's polarizability theory [52] under the conditions that:

- 1) the frequency of the incident radiation is much larger than the frequency of any vibration-rotation transition of the molecule,
- 2) the frequency of the incident radiation is much smaller than any electronic transition frequency of the molecule,
- 3) the ground electronic state of the molecule is not degenerate.

These conditions, which exclude resonant scattering processes, are well satisfied for typical laser frequencies used in lidar and for the atmospheric molecules of interest.

The characteristic molecular parameters that determine the cross sections of a diatomic molecule are the mean polarizability  $a$  and the anisotropy of the polarizability  $\gamma$  and their derivatives with respect to the normal coordinate of vibration at the equilibrium position,  $a'$  and  $\gamma'$ .  $a^2$  and  $\gamma^2$  are invariants of the molecule's polarizability tensor. The derivatives  $a'$  and  $\gamma'$  characterize the change of the polarizability properties with changing distance between the atoms during vibration. From these parameters we get the differential cross section for Rayleigh backscattering,

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{Ray}} = k_{\tilde{\nu}} \tilde{\nu}_1^4 \left(a^2 + \frac{7}{180}\gamma^2\right), \quad (9.5)$$

the differential cross section for pure rotational Raman backscattering,

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{RR}} = k_{\tilde{\nu}} \tilde{\nu}_1^4 \left(\frac{7}{60}\gamma^2\right), \quad (9.6)$$

the differential cross section for Stokes vibration–rotation Raman backscattering,

$$\begin{aligned} \left(\frac{d\sigma}{d\Omega}\right)_{\text{VRR}}^{\text{Stokes}} &= k_{\tilde{\nu}} (\tilde{\nu}_1 - \tilde{\nu}_{\text{vib}})^4 \frac{b_v^2}{[1 - \exp(-hc_0\tilde{\nu}_{\text{vib}}/k_{\text{B}}T)]} \\ &\times \left(a'^2 + \frac{7}{45}\gamma'^2\right), \end{aligned} \quad (9.7)$$

and the differential cross section for anti-Stokes vibration–rotation Raman backscattering,

$$\begin{aligned} \left(\frac{d\sigma}{d\Omega}\right)_{\text{VRR}}^{\text{anti-Stokes}} &= k_{\tilde{\nu}} (\tilde{\nu}_1 + \tilde{\nu}_{\text{vib}})^4 \frac{b_v^2}{[\exp(hc_0\tilde{\nu}_{\text{vib}}/k_{\text{B}}T) - 1]} \\ &\times \left(a'^2 + \frac{7}{45}\gamma'^2\right), \end{aligned} \quad (9.8)$$

with the square of the zero-point amplitude of the vibrational mode

$$b_v^2 = \frac{h}{8\pi^2 c_0 \tilde{\nu}_{\text{vib}}}, \quad (9.9)$$

and

$$k_{\tilde{\nu}} = \frac{\pi^2}{\epsilon_0^2}, \quad (9.10)$$

with the permittivity of vacuum  $\epsilon_0$ . There are several remarks to be made in conjunction with these equations.

1) The cross sections in the given form hold for an assembly of molecules. Under atmospheric conditions, most of the molecules will be in their vibrational ground state with quantum number  $v = 0$ . However, there will be some population of higher vibrational energy levels with  $v = 1, 2, \dots$ . The fraction of molecules in each vibrational state is calculated from the actual absolute temperature  $T$  after the Boltzmann distribution law with the Boltzmann constant  $k_B$ . In this way, we obtain the terms with the exponential function in square brackets in Eqs. (9.7) and (9.8) for the vibration–rotation transitions. From the ratio of these equations the intensity ratio of Stokes to anti-Stokes vibration–rotation Raman scattering is obtained as

$$\frac{(\mathrm{d}\sigma/\mathrm{d}\Omega)_{\mathrm{VRR}}^{\mathrm{Stokes}}}{(\mathrm{d}\sigma/\mathrm{d}\Omega)_{\mathrm{VRR}}^{\mathrm{anti-Stokes}}} = \frac{(\tilde{\nu}_1 - \tilde{\nu}_{\mathrm{vib}})^4}{(\tilde{\nu}_1 + \tilde{\nu}_{\mathrm{vib}})^4} \exp(hc_0\tilde{\nu}_{\mathrm{vib}}/k_B T). \quad (9.11)$$

From this equation, we see that under atmospheric conditions the anti-Stokes vibration–rotation Raman bands have 3–6 orders less intensity than the corresponding Stokes bands. In a similar way, we can calculate the intensity ratios between Rayleigh and Raman scattered radiation or between rotational and vibration–rotation Raman scattered radiation from Eqs. (9.5)–(9.8).

2) Equations (9.5)–(9.8) hold for linearly polarized or unpolarized incident light and the polarization-independent observation of the complete backscatter signal. The equations can be transferred to other polarization configurations with the help of the Reference Tables of Ref. [53], the Central Reference Section of Ref. [55], or Ref. [62].

3) Equations (9.6)–(9.8) give the backscatter cross sections for the complete rotational and vibration–rotation Raman bands. The contributions from the different branches are obtained by appropriate separation of the last terms in Eqs. (9.5)–(9.8), i.e.,

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)^{\mathrm{center}} \sim \left(a_l^2 + \frac{7}{180}\gamma_l^2\right) \text{ and } \left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)^{\mathrm{wings}} \sim \left(\frac{7}{60}\gamma_l^2\right), \quad (9.12)$$

with  $a_l^2 = a^2, a'^2$  and  $\gamma_l^2 = \gamma^2, \gamma'^2$ . The superscript *center* stands for either the Rayleigh line or the Q branch whereas *wings* describes the rotational side bands (O and S branches, see Fig. 9.1). We now see the equivalence between Rayleigh and rotational Raman scattering, on the one hand, and vibration–rotation Raman scattering, on the other hand.



Often the sum of Eqs. (9.5) and (9.6) is given as Rayleigh backscatter cross section, i.e., the contribution of rotational Raman scattering is added to the so-called Cabannes line [Eq. (9.5),  $\Delta v = 0$ ,  $\Delta J = 0$ ] and both together are referred to as Rayleigh scattering. Such a definition makes sense if the backscatter signal is detected with low spectral resolution, so that the rotational Raman lines contribute to the elastic backscatter signal measured with lidar.

4) If we want to calculate intensities of single Raman lines within the different branches, we need the population distribution for the initial rotational energy levels. Because the energy difference between the rotational levels is much smaller than between the vibrational levels, higher rotational energy levels of the vibrational ground state are well populated under atmospheric conditions. The population distribution follows again from the Boltzmann distribution law. In addition, we have to consider the degeneracy  $2J + 1$  of the initial rotational energy level  $J$ , the nuclear-spin degeneracy  $2I + 1$ , and the nuclear-spin statistical weight factor  $g_N$ . The differential backscatter cross section for single lines can then be written as

$$\left(\frac{d\sigma}{d\Omega}\right)_J^{\text{RR,VRR}} = k_{\tilde{\nu}}(\tilde{\nu}_1 \mp |\Delta\tilde{\nu}|)^4 \frac{g_N \Phi_J}{Q} \exp\left[-\frac{B_i hc_0 J(J+1)}{k_B T}\right], \quad (9.13)$$

with the actual frequency shift of the line  $|\Delta\tilde{\nu}|$ .  $B_i$  is the rotational constant of the initial vibrational state, i.e.,  $B_i = B_0$  for rotational and Stokes vibration–rotation lines and  $B_i = B_1$  for anti-Stokes vibration–rotation lines.  $Q \approx k_B T / 2hc_0 B_0$  is the state sum or partition function.  $N_2$  and  $O_2$  have nuclear spins of  $I = 1$  and  $I = 0$ , respectively, which leads to different nuclear spin statistics. In addition, for homonuclear molecules  $g_N$  depends on the initial rotational state  $J$ , and we obtain for  $N_2$

$$g_N = \begin{cases} 6 & \text{for } J \text{ even} \\ 3 & \text{for } J \text{ odd} \end{cases} \quad (9.14)$$

and for  $O_2$

$$g_N = \begin{cases} 0 & \text{for } J \text{ even} \\ 1 & \text{for } J \text{ odd.} \end{cases} \quad (9.15)$$

From this condition it follows that every second Raman line of  $O_2$  is missing and that the Raman lines of  $N_2$  show an alternating intensity. The function  $\Phi_J$  contains the Placzek–Teller factors, the degeneracy

$2J + 1$ , and the factor with the molecular constants for the specific observational geometry. In our case (see remark 2) we have to insert for Stokes rotational lines

$$\Phi_J = \frac{7(J+1)(J+2)}{30(2J+3)}\gamma^2, \quad J = 0, 1, 2, \dots, \quad (9.16)$$

for anti-Stokes rotational lines

$$\Phi_J = \frac{7J(J-1)}{30(2J-1)}\gamma^2, \quad J = 2, 3, 4, \dots, \quad (9.17)$$

for Stokes vibration–rotation lines of the Q branch

$$\Phi_J = \frac{b_v^2(2J+1)}{[1 - \exp(-hc_0\tilde{\nu}_{\text{vib}}/k_B T)]} \left[ a'^2 + \frac{7J(J+1)}{45(2J+3)(2J-1)}\gamma'^2 \right],$$

$$J = 0, 1, 2, \dots, \quad (9.18)$$

for Stokes vibration–rotation lines of the O branch

$$\Phi_J = \frac{b_v^2}{[1 - \exp(-hc_0\tilde{\nu}_{\text{vib}}/k_B T)]} \frac{7J(J-1)}{30(2J-1)}\gamma'^2, \quad J = 2, 3, 4, \dots, \quad (9.19)$$

and for Stokes vibration–rotation lines of the S branch

$$\Phi_J = \frac{b_v^2}{[1 - \exp(-hc_0\tilde{\nu}_{\text{vib}}/k_B T)]} \frac{7(J+1)(J+2)}{30(2J+3)}\gamma'^2,$$

$$J = 0, 1, 2, \dots \quad (9.20)$$

In Eqs. (9.13)–(9.20)  $J$  is the rotational quantum number of the initial state. Some authors prefer to use the rotational quantum number  $J''$  of the lower energy level involved in the transition instead, which leads to a modification of the equations for anti-Stokes transitions. The latter notation has the advantage that rotational lines of the Stokes and anti-Stokes branches which have the same absolute frequency shifts, i.e., which are symmetric in the spectrum with respect to  $\tilde{\nu}_1$ , are identified by the same  $J$  number.

5) The constants  $a^2$ ,  $\gamma^2$ ,  $a'^2$ , and  $\gamma'^2$  have been determined from different experimental and theoretical approaches for various molecular species. Using values from the literature, one has to be careful as to the specific definition of the polarizability properties (relations are given in Ref. [53], p. 126–127). In older literature, cgs units are used, and the

values have to be either multiplied by  $(4\pi\epsilon_0)^2$  before they can be used in the equations given above or the constant  $k_{\bar{\nu}}$  in the equations has to be replaced by  $(2\pi)^4$ . The molecular constants show a slight dispersion, i.e., they depend on the wavelength of the incident radiation. For  $a^2$  the dispersion can be calculated from the corresponding dispersion of the refractive index  $n$ , since

$$a = \frac{2\epsilon_0(n-1)}{N}, \quad (9.21)$$

with the molecule number density  $N$ . We find that  $a^2$  decreases by about 5–10% if the incident wavelength increases from 300 to 600 nm, i.e., the dispersion is of the order of  $10^{-3}$  to  $10^{-4} \text{ nm}^{-1}$  and thus negligible for our applications. The same holds for the other constants [63, 64]. Table 9.3 summarizes the molecular constants for the calculation of Raman scattering cross sections of nitrogen and oxygen obtained experimentally at wavelengths between 400 and 500 nm [63, 65, 66]. The factor  $(4\pi\epsilon_0)^2$  is indicated separately in order to show the original numbers from the literature.

The third atmospheric molecule of interest,  $\text{H}_2\text{O}$ , is a nonlinear, asymmetric top molecule and the calculation of its Raman spectrum is much more complicated. A recent paper [67] deals with the calculation of the vibration–rotation Raman scattering cross sections of  $\text{H}_2\text{O}$  that follow from the symmetric and antisymmetric O–H stretching vibrations. A table with the molecular constants for more than 7000 vibration–rotation transitions is given. These data were used in the calculation of an atmospheric backscatter spectrum shown in Fig. 9.2 for an incident wavelength of 355 nm. For Rayleigh scattering and for nitrogen and oxygen Raman scattering the positions and intensities of the lines were calculated from the equations given above. In addition, the Raman bands of liquid water and ice are shown in Fig. 9.2 on an arbitrary intensity scale [68–70].

**Table 9.3.** Molecular constants for the calculation of Raman scattering cross sections of nitrogen and oxygen

| Gas          | $a^2$<br>$\text{m}^6/(4\pi\epsilon_0)^2$ | $\gamma^2$<br>$\text{m}^6/(4\pi\epsilon_0)^2$ | $a'^2$<br>$\text{m}^4/\text{kg}/(4\pi\epsilon_0)^2$ | $\gamma'^2$<br>$\text{m}^4/\text{kg}/(4\pi\epsilon_0)^2$ |
|--------------|------------------------------------------|-----------------------------------------------|-----------------------------------------------------|----------------------------------------------------------|
| $\text{N}_2$ | $3.17 \times 10^{-60}$                   | $0.52 \times 10^{-60}$                        | $2.62 \times 10^{-14}$                              | $4.23 \times 10^{-14}$                                   |
| $\text{O}_2$ | $2.66 \times 10^{-60}$                   | $1.26 \times 10^{-60}$                        | $1.63 \times 10^{-14}$                              | $6.46 \times 10^{-14}$                                   |

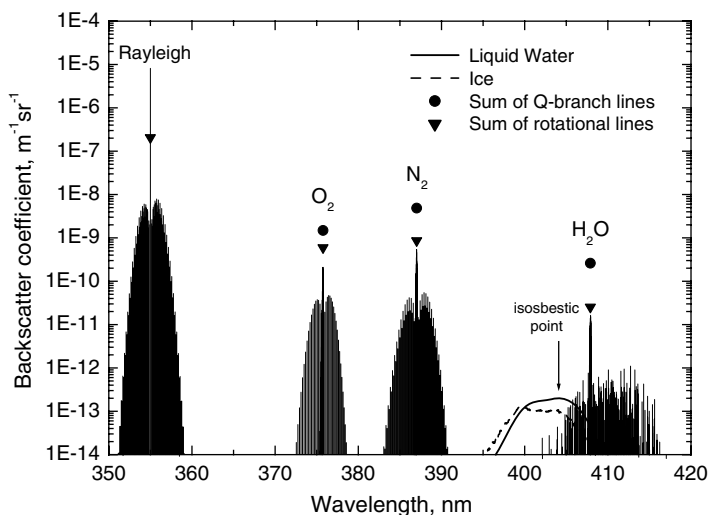


**Fig. 9.2.** Raman backscatter spectrum of the atmosphere for an incident laser wavelength of 355 nm, normal pressure, a temperature of 300 K, an  $\text{N}_2$  and  $\text{O}_2$  content of 0.781 and 0.209, respectively, and a water-vapor mixing ratio of 10 g/kg. The curves for liquid water and ice are arbitrarily scaled. The isosbestic point is discussed in Subsection 9.5.2.

## 9.3 Technical Requirements

The low intensity of the Raman backscatter signals calls for a specific technical lidar setup. A high-power laser transmitter and a highly efficient receiving and detection system are required. Figure 9.3 shows schematically the setup of an aerosol and water-vapor Raman lidar with one emitted wavelength. The lidar consists of a laser, a beam-expanding and transmitter optics, a receiver telescope with field stop, three detection channels for the measurement of the water-vapor and nitrogen Raman signals and the elastic signal, and a data-acquisition and computer unit. The specific requirements for each of the components are explained in the following.

### 9.3.1 Laser

As mentioned previously, the Raman lidar technique does not require specific emission wavelengths or high spectral purity of the emitted laser light, but a high average laser power and a preferred emission wavelength between 320 and 550 nm. In the beginning of atmospheric Raman lidar observations the nitrogen laser at 337 nm and the ruby laser

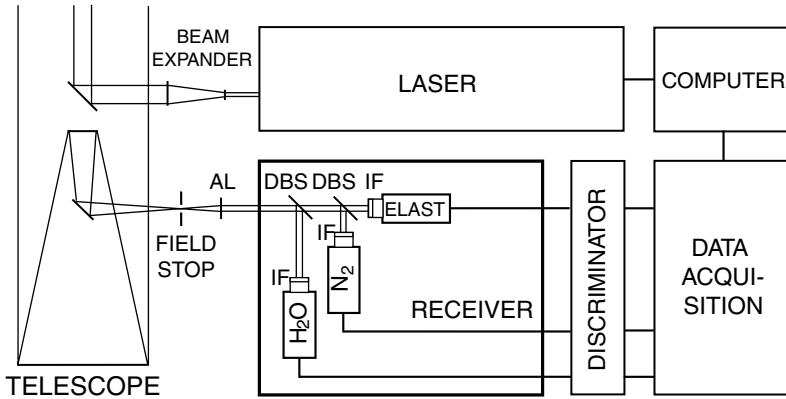


**Fig. 9.3.** Typical setup of a water-vapor and aerosol Raman lidar. AL – achromatic lens, IF – interference filter, DBS – dichroic beam splitter.

at its fundamental of 694 nm or frequency-doubled to 347 nm were used [1–6, 71]. Because of the relatively low average power of these lasers, early atmospheric Raman measurements were limited to ranges of about 2 km. Pulsed lasers with high average power in the visible and ultraviolet spectral region became available during the 1980s. UV excimer lasers, i.e., XeCl and XeF lasers at 308 and 351 nm, and frequency-tripled Nd:YAG lasers at 355 nm were used first to obtain Raman scattering up to the middle [72] and upper troposphere [9, 10, 73, 74]. Nowadays, the Nd:YAG laser is the workhorse in the Raman lidar field [11, 13, 16, 75–77]. Its primary wavelength of 1064 nm is converted to 532 and 355 nm by frequency-doubling and frequency-tripling techniques. Laser pulse repetition rates of 20 to 50 s<sup>-1</sup> and pulse energies of 0.5 to 1.5 J at the primary wavelength, resulting in an average power >10 W, are typically used. For the same average power, a higher pulse energy at a lower pulse repetition rate is to be preferred because the signal-to-noise ratio of the measurements, especially at daytime, is improved in this way.

### 9.3.2 Beam Expander

The typical laser beam divergence of  $\sim 1$  mrad must be reduced by appropriate beam expansion in order to allow for a narrow telescope field of view, which again will help to suppress background light and thus increase the signal-to-noise ratio. Beam expansion by a factor  $n$  reduces the divergence by the same factor. A typical beam-expansion factor is  $n = 10$ . Reduction of the divergence to less than 0.1 mrad does

not further improve the characteristics of ground-based systems because turbulence in the lower troposphere leads to an effective beam divergence of about that figure.

Mirror or lens telescopes can be used as beam expanders. A laser surface coating for high reflectivity at the emission wavelength in the case of the mirror reflectors is necessary. Lenses need an anti-reflection coating to avoid back-reflection from the surfaces into the laser cavity. Achromatic optics is necessary if light is emitted at more than one wavelength.

### 9.3.3 Telescope

Receiver telescopes of diameters of the order of 0.5–1 m are typically used in Raman lidar systems. Different configurations, mainly of Cassegrainian type, with ratios of effective focal length to primary aperture of  $f/(3 \dots 6)$ , are applied [9, 10, 13, 76]. Coaxial (laser beam is on the optical axis of the telescope) as well as biaxial setups (laser beam is off the optical axis of the telescope and possibly tilted against it) are realized. A field stop in the focal point of the telescope determines the receiver field of view (RFOV). The RFOV is normally a factor of 2–10 larger than the laser beam divergence. A compromise must be found between a small RFOV necessary for high background suppression and a larger RFOV for stable adjustment of the laser beam within the RFOV and for a sufficient signal intensity from short distances.

In the telescope design care must be taken concerning the laser-beam RFOV overlap factor, which is influenced by the laser beam divergence, the RFOV, the imaging properties of the telescope (depth of focus), geometric shadows of the secondary mirror and of mountings within the RFOV, and the relative tilt angle between the laser beam and the optical axis of the telescope. For Raman measurements of water vapor and other gases, particle backscatter coefficient, or temperature, which are calculated from a signal ratio (see Section 9.4 and Chapters 4 and 10), it is sufficient to realize equivalent optical paths of the two signals in the receiver, because then the overlap factor cancels out. Extinction profiling requires an overlap factor of 1 or the exact knowledge of the overlap factor *vs* distance, which is often not given at short range up to 1 km distance from the lidar [78]. Therefore, in high-performance systems a second, small-diameter telescope with larger RFOV is used to cover the near range. The use of a separate near-field telescope helps to keep the large telescope optimized for good background suppression. In addition,

the dynamic range of the system is increased because the lidar signal is divided into a short-range and a long-range part. The shortcoming of this approach is that a second receiver chain has to be implemented which increases complexity and cost of the system.

#### 9.3.4 Receiver Optics

The receiver optics behind the telescope must be optimized for high transmission of the Raman signals. In addition, elastically backscattered light in the Raman channels must be suppressed. In the setup shown in Fig. 9.3 a suppression factor of  $10^8$  and  $10^{10}$  is necessary in the vibration–rotation nitrogen and water-vapor Raman channels, respectively. Dichroic beam splitters and interference filters are mainly used for this purpose [11, 13, 16, 76]. Dichroic beam splitters reflect light of a certain wavelength range with high efficiency whereas they transmit light of other spectral regions. Interference filters with a bandwidth of  $<0.5$  nm, a peak transmission of 50–70%, and an excellent out-of-band suppression have become available during the past decade. They are one of the reasons why nowadays the Raman lidar technique turns out to be very stable and robust and a good candidate for routine and automated observations.

Grating monochromators can be applied for the wavelength separation as well, especially if high spectral resolution is required as in the case of Raman temperature measurements (see Chapter 10). A double-grating setup or a combination of a grating with filters is necessary to sufficiently suppress the elastically scattered light [16, 37].

#### 9.3.5 Detectors and Data Acquisition

Photomultipliers in photon-counting mode are typically used in Raman lidar systems. High quantum efficiency and low noise are required. In the ultraviolet region, a quantum efficiency of 25% and a dark count rate of  $<5$  s<sup>-1</sup> can be achieved. The detector output pulses can be pre-amplified before discrimination and registration. Counters on the basis of multichannel scalers are used to acquire the signals. To start data acquisition precisely at the time when the laser pulse enters the atmosphere, a trigger signal from a detector (fast photodiode) that senses a fraction of the outgoing pulse can be used. The typical time resolution or window length of data acquisition is  $\sim 100$  ns which corresponds to a range resolution of 15 m. Typical averaging time for the raw signals is 10–30 s.

(The signals are usually further averaged in time and space during data evaluation.) The number of counts per range gate and unit time is finally stored on a computer.

The whole detector chain from the photomultiplier to the data acquisition system should allow for count rates  $\geq 250$  MHz or a dead time of  $\leq 4$  ns. Then, assuming Poisson statistics for the temporal distribution of backscattered photons, dead-time effects will not significantly influence count rates  $\leq 10$  MHz. Therefore, the maximum count rate must be limited to about that figure by appropriate signal reduction, e.g., by inserting neutral-density filters. Dead-time correction is possible up to about 80 MHz, if the dead-time behavior of the system is exactly known [79, 80]. Under daylight conditions and in the near range, the Raman signals allow analog detection as well. A combination of analog and photon-counting detection can help to increase the system's dynamic range [47].

## 9.4 Measurement of Water Vapor

In the following, we describe the data evaluation procedure for Raman measurements of gas concentrations. In principle, this procedure is valid for the measurement of any Raman-active gas with sufficiently high atmospheric concentration. As mentioned in the Introduction, we will focus the discussion on the detection of water vapor as the most important observable in this context.

### 9.4.1 Mixing Ratio

The Raman signal  $P_R(z)$  from distance  $z$  measured with lidar at the Raman wavelength  $\lambda_R$  is described by the Raman lidar equation, which can be written as

$$P_R(z) = \frac{K_R O(z)}{z^2} \beta_R(z) \exp \left\{ - \int_0^z [\alpha_0(\zeta) + \alpha_R(\zeta)] d\zeta \right\}. \quad (9.22)$$

$O(z)$  is the factor describing the overlap between the laser beam and the RFOV and is equal to 1 for heights above which the laser beam is completely imaged onto the photomultiplier cathode.  $K_R$  comprises all range-independent system parameters such as telescope area, receiver transmission and detection efficiency.  $\beta_R$  is the Raman backscatter cross section, and  $\alpha_0(z)$  and  $\alpha_R(z)$  describe the extinction of light on the way



from the lidar to the backscatter region and on the way back to the lidar after the Raman scattering process, respectively. The latter two parameters include light extinction due to Rayleigh scattering and to scattering and absorption by aerosol particles. Molecular absorption can also contribute to this term (see Subsection 9.5.1). The Raman lidar equation (9.22) differs from the usual lidar equation (see Chapter 1) only in the way that 1) the Raman backscatter coefficient

$$\beta_R = N_R(z) \frac{d\sigma(\pi)}{d\Omega} \quad (9.23)$$

is given by the molecule number density  $N_R(z)$  of the Raman-active gas and the differential Raman cross section for the backward direction  $d\sigma(\pi)/d\Omega$  (see Section 9.2) and that 2) light extinction on the way back from the scattering volume must be considered at the Raman-shifted wavelength  $\lambda_R$ . Depending on the spectral resolution of the receiver,  $\beta_R$  and thus  $P_R(z)$  can describe the scattering according to a single Raman line, a group of lines, or a whole rotational or vibration–rotation Raman band (see Section 9.2).

The Raman method for gas-concentration measurements, e.g., of water vapor, makes use of two Raman signals, one of which is the return signal  $P_R$  from the gas of interest, usually the Stokes vibration–rotation Raman band or a part of it, and the other one is the signal  $P_{\text{Ref}}$  of a reference gas. The Stokes vibration-rotation Raman band of  $\text{N}_2$  is typically used as the reference signal. However, the pure rotational Raman band of  $\text{N}_2$  and/or  $\text{O}_2$  can serve for this purpose as well, if a temperature-insensitive part of the band is chosen and elastic scattering is suppressed sufficiently well [49].

By forming the signal ratio  $P_R/P_{\text{Ref}}$  and rearranging the resulting equation, we obtain the mixing ratio of the gas relative to dry air:

$$m(z) = C \frac{P_R(z)}{P_{\text{Ref}}(z)} \frac{\exp[-\int_0^z \alpha_{\text{Ref}}(\zeta) d\zeta]}{\exp[-\int_0^z \alpha_R(\zeta) d\zeta]}, \quad (9.24)$$

with the calibration constant  $C$ . Equation (9.24) assumes identical overlap factors and range-independent Raman backscatter cross sections for the two signals. The difference between the atmospheric transmission at  $\lambda_R$  and the one at  $\lambda_{\text{Ref}}$  is mainly caused by Rayleigh scattering and can easily be corrected by using standard atmosphere profiles of temperature and pressure, or, if available, actual radiosonde data. The meteorological data yield the molecule number density. The Rayleigh scattering cross

sections are constant and taken from the literature [81]. Differences in the transmissions at the two Raman wavelengths caused by wavelength-dependent particle extinction are found to be negligible in most cases (see Subsection 9.4.3).

## 9.4.2 Calibration

Equation (9.24) requires the calibration of the relative measure of the gas concentration. The calibration constant  $C$  accounts for the ratio  $M_R/M_{\text{air}}$  of the molecule masses of the investigated gas and of air, for the ratio  $N_{\text{Ref}}/N_{\text{air}}$  of the molecule number densities of the reference gas and of air, for the transmission and detection efficiency ratio of the lidar system for the two signals, and for the ratio of the *effective* Raman backscatter cross sections of the two gases. We introduce the term *effective* cross section because, particularly when using narrow-bandwidth filters for the selection of the Raman signals, we have to take the average cross section over the spectrum of the observed Raman band, weighted with the instrument's transmission function. In this context we also have to consider a possible temperature, and thus range, dependence of the calibration constant due to a temperature dependence of the signal intensity distribution in the wings of the Raman bands [82, 83].

The calibration constant can be determined in different ways. Most often the lidar measurement is calibrated against an *in situ* observation of the same quantity. In the case of water vapor, it is usually the measurement of the mixing-ratio profile with an accompanying radiosonde. This procedure makes the lidar measurement dependent on the measurement accuracy of a second instrument. The accuracy of lidar calibration with radiosondes has been discussed extensively in the literature. We refer to Ref. [82] for an overview. Several other attempts have been made to develop calibration methods especially for water-vapor observations. For example, the calibration of the range-integrated profile against an atmospheric column value measured with a microwave radiometer has been suggested [84].

In principle, an independent calibration of the system is possible by measuring or calculating the relevant system parameters. The use of two identical filters to obtain nitrogen Raman scattering in both channels can help to estimate their transmission efficiency ratio [85]. Sherlock et al. [82] used diffuse daylight to determine the transmission ratio of the two instrument channels and calculated the ratio of the effective cross sections. The system's short-term and long-term stability together

with the uncertainties inherent in any of the calibration methods lead to the conclusion that calibration errors remain on the order of 5% in any case [82].

### 9.4.3 Errors

#### Statistical Errors

The error of Raman lidar measurements is usually dominated by statistical noise. We can assume that the detected photons follow a Poisson distribution, so that the statistical error  $\Delta P_i = \sqrt{P_i}$  of each signal is calculated from the number of acquired photon counts  $P_i$ , with  $i = \text{R, Ref.}$  Each signal is the sum of counts from detected Raman-backscattered photons and from sky background photons and electronic noise. The background signal is usually determined at the far end of the lidar range from which no Raman backscattered photons are detected any more, and then subtracted. Both signal noise and background noise contribute to the statistical error of the mixing ratio, which is obtained by applying the law of error propagation to Eq. (9.24).

The achievable measurement accuracy in Raman lidar depends primarily on the system parameters (laser power, telescope diameter), but also on the actual measurement conditions (gas concentration, aerosol extinction). Signals ought to be averaged in time and space until the signal error is <20% before the data evaluation procedures are applied and total errors are calculated according to the rules of error propagation.

#### Systematic Errors

Systematic errors mainly arise from the calibration procedure as discussed before. If an accompanying radiosonde is used for calibration, the calibration error can be estimated, e.g., by calculating the standard deviation of the calibration constant from a fit of the relative mixing ratio ( $C = 1$ ) to the radiosonde profile over a certain height range (see Subsection 9.4.4). Furthermore, the variation of the calibration constant by using multiple sondes at different times can be investigated.

As mentioned before, the possible temperature dependence of the effective Raman backscatter cross sections due to narrow filter bandwidths and the resulting altitude dependence of the calibration factor should be investigated for each individual lidar system. Whiteman [80]

found a change in the water-vapor backscatter cross section of 7% for a filter bandwidth of 0.34 nm and a temperature variation between 200 and 300 K.

Other systematic errors follow from the correction of the differential atmospheric transmission for the two signals because of the different extinction on the way back from the scattering volume to the lidar system [exponential terms in Eq. (9.24)]. The correction of Rayleigh scattering is straightforward as already mentioned and errors due to an uncertainty in the applied temperature and pressure profiles are negligible, even if no accompanying radiosonde measurement is available and standard-atmosphere conditions are assumed.

Differential aerosol extinction plays a role for large aerosol optical depths only. The extinction profile is determined from the Raman reference profile for heights for which the overlap factor is 1 or an overlap correction can be applied (see [42, 78] and Chapter 4). For lower heights a linear extrapolation of the extinction profile can be used. For the calculation of the differential aerosol extinction a parameter  $k$ , which accounts for the wavelength dependence of aerosol extinction, must be assumed. This parameter, in the literature often referred to as the Ångström exponent and defined as  $k = \ln(\alpha_1/\alpha_2)/\ln(\lambda_2/\lambda_1)$ , with the extinction coefficients  $\alpha_1$  and  $\alpha_2$  at the wavelengths  $\lambda_1$  and  $\lambda_2$ , respectively, may vary typically between 0 for marine aerosol or large dust particles and 2 for small industrial pollution particles. Airmass characterization may thus help to estimate  $k$ . However, if  $k = 1$  is assumed in general and the true  $k$  is 0 or 2, the resulting error of the mixing ratio from the transmission correction is <3% for aerosol optical depths <0.5. The error may reach values up to 10% if the aerosol optical depth is as high as 2 [9, 16, 86]. Advanced Raman lidar systems measure aerosol extinction at more than one wavelength simultaneously, so that the Ångström exponent can even be determined with the same system [87, 88].

Differential ozone absorption must be considered in the transmission correction for laser wavelengths <300 nm only. If, e.g., the XeCl excimer laser at 308 nm is used for water-vapor measurements, the detection wavelengths for water vapor and nitrogen are at 347 and 332 nm, respectively, and the resulting error by neglecting differential ozone absorption is <1% [86]. In the solar-blind region, the accompanying measurement of ozone profiles, e.g., by applying the ozone Raman DIAL technique (see Subsection 9.5.1) is advisable.

### 9.4.4 Example

Figure 9.4 shows an example of a water-vapor measurement. The measurement was made with the Raman lidar of the Institute for Tropospheric Research, Leipzig, Germany, on April 23, 2002, between 1930 and 2132 UTC [16]. The mixing-ratio profile was calibrated against the profile from a Vaisala RS-80 radiosonde launched at 2010 UTC at the lidar site. The calibration constant was determined in the height range from 0 to 6.5 km as  $0.00877 \pm 0.00054$ , i.e., with an error of about 6%. The right panel of Fig. 9.4 shows the variability of the calibration constant which is mainly attributed to the atmospheric variability during the time of the measurement. The variations are highest in the lower 4 km of the atmosphere. Above 6.5 km the typical underestimation of the water-vapor content for temperatures  $< -40^\circ\text{C}$  with the radiosonde can be seen. Mixing ratios  $> 1$  g/kg are shown on a linear, those  $< 1$  g/kg on a logarithmic scale in the left panel of Fig. 9.4. The Raman measurements of the water-vapor mixing ratio typically cover values over three orders of magnitude. The error bars in the figure account for statistical noise and the systematic calibration error.

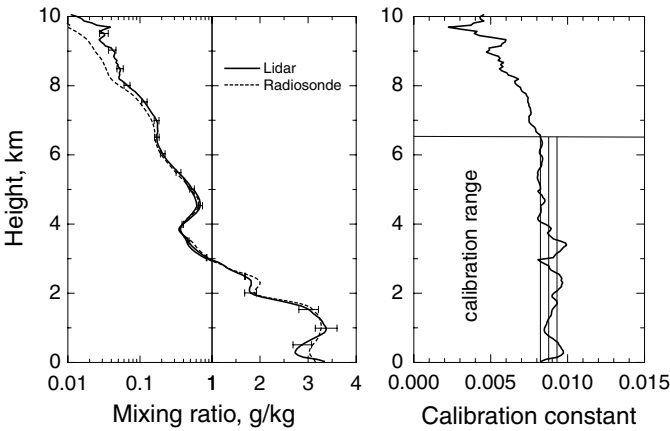


**Fig. 9.4.** Left: Water-vapor mixing-ratio profile determined with Raman lidar and an accompanying radiosonde. The temporal resolution of the lidar measurement is 2 h, the spatial resolution is 120 m from 0 to 3 km, 480 m from 3 to 5 km and 1200 m above. Right: Calibration constant defined as the ratio of the water-vapor mixing ratio determined with radiosonde and the one obtained with the uncalibrated lidar [ $C = 1$  in Eq. (9.24)], mean value and standard deviation for the height range 0–6.5 km.

## 9.5 Other Applications

### 9.5.1 Ozone Raman DIAL

Ozone Raman DIAL is based on the differential-absorption lidar or DIAL method, i.e., it makes use of two backscatter signals one of which is more strongly absorbed by ozone than the other (see Chapter 7). Unlike in conventional DIAL, the two signals are not elastically backscattered by molecules and particles, but Raman backscattered by molecules of nitrogen and/or oxygen alone. The advantage of Raman DIAL against conventional DIAL is that the error term due to differential backscattering disappears. Conventional DIAL ozone measurements in regions with inhomogeneous aerosol load are hard to carry out because of this error, particularly if the two wavelengths are separated widely as it is the case for stratospheric lidar systems and also for many tropospheric systems working in the solar-blind region.

After the eruption of Mt. Pinatubo in 1991, the stratosphere was globally contaminated with sulfuric-acid particles for several years and the conventional stratospheric ozone DIAL method could not be applied any more. At that time McGee et al. [44, 89] suggested using two nitrogen Raman signals for the measurement of differential ozone absorption. They used two laser wavelengths of a conventional DIAL system, 308 nm from a XeCl laser and 351 nm from a XeF laser, and measured the Stokes vibration–rotation nitrogen Raman bands at 332 and 382 nm as the on-line and off-line signals, respectively. With a combination of a XeCl and a frequency-tripled Nd:YAG laser (355 nm; N<sub>2</sub> VRR at 387 nm) Reichardt et al. [45, 90] applied the technique to the measurement of ozone in cirrus clouds. Later, Reichardt et al. [46] showed that the method also works with a single XeCl laser, if the pure rotational signal of nitrogen and oxygen is used in conjunction with a vibration–rotation Raman band of oxygen or nitrogen. They called this technique RVR Raman DIAL. This approach not only reduces the complexity of the lidar system, but the sensitivity of the Raman DIAL method to multiple scattering effects in clouds as well [91], because of the smaller wavelength difference between the on- and the off-line signal.

Figure 9.5 shows the ozone absorption cross section in the 250-to-350-nm spectral region of the Hartley band [92] together with the wavelengths  $\lambda > 300$  nm used for the ozone Raman DIAL measurements in the upper troposphere and stratosphere described above. Also shown are wavelengths that can be used for ozone Raman DIAL applications in

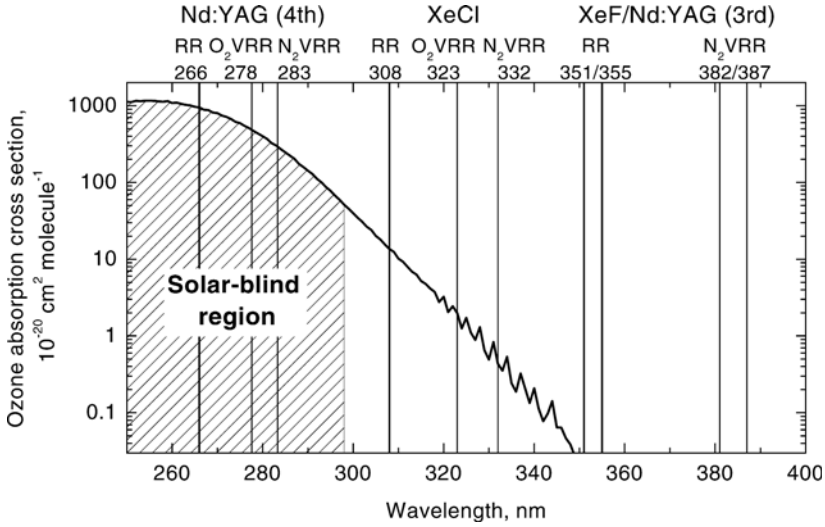


**Fig. 9.5.** Ozone absorption cross section and wavelengths applied in the ozone Raman DIAL technique for upper tropospheric and stratospheric measurements ( $\lambda > 300$  nm) and for boundary-layer measurements in the solar-blind region ( $\lambda < 300$  nm).

the solar-blind region (shaded area). The fourth harmonic of the Nd:YAG laser is a typical laser source here and the vibration–rotation Raman lines of oxygen and nitrogen at 278 and 283 nm are then the on-line and off-line signals, respectively. Measurements of ozone concentrations in the planetary boundary layer up to about 2 km height can be made with this technique independent of the time of the day [29, 93, 94].

The two reference signals used in the Raman DIAL method may be written as  $P_{\text{Ref}}^{\text{on}}$  and  $P_{\text{Ref}}^{\text{off}}$ . By differentiation of the logarithm of the ratio of these two signals, we obtain the ozone molecule number density

$$N_{\text{O}_3}(z) = \frac{1}{\Delta\sigma_{\text{abs}}(T)} \left( \frac{d}{dz} \ln \frac{P_{\text{Ref}}^{\text{on}}(z)}{P_{\text{Ref}}^{\text{off}}(z)} - \Delta\alpha_{\text{aer}}(z) - \Delta\alpha_{\text{mol}}(z) \right). \quad (9.25)$$

Here we must consider the extinction because of ozone absorption,  $\alpha_{\text{abs}} = N_{\text{O}_3} \sigma_{\text{abs}}$ , with the ozone absorption cross section  $\sigma_{\text{abs}}$ , in the exponential term of the Raman lidar equation (9.22). The ozone absorption cross section  $\sigma_{\text{abs}}$  is a function of temperature and therefore depends on height  $z$ . For a Raman DIAL system with two emission wavelengths, the three  $\Delta$  expressions consist of four terms each,

$$\Delta\xi = \xi(\lambda_0^{\text{on}}) + \xi(\lambda_{\text{R}}^{\text{on}}) - \xi(\lambda_0^{\text{off}}) - \xi(\lambda_{\text{R}}^{\text{off}}), \quad (9.26)$$

with  $\xi = \sigma_{\text{abs}}, \alpha_{\text{mol}}, \alpha_{\text{aer}}$ .  $\lambda_0^{\text{on}}$  and  $\lambda_0^{\text{off}}$  are the laser wavelengths and  $\lambda_{\text{R}}^{\text{on}}$  and  $\lambda_{\text{R}}^{\text{off}}$  the wavelengths of the vibration–rotation bands. For RVR Raman DIAL the equation reduces to

$$\Delta\xi = \xi(\lambda_{\text{R}}^{\text{on}}) - \xi(\lambda_{\text{R}}^{\text{off}}), \quad (9.27)$$

with the wavelengths  $\lambda_{\text{R}}^{\text{on}}$  and  $\lambda_{\text{R}}^{\text{off}}$  of the pure rotational and the vibration–rotation band, respectively.

The calculation of the differential Rayleigh scattering and the differential aerosol extinction is done in the same way as discussed in Subsection 9.4.3 for water vapor, i.e., Rayleigh scattering is calculated from temperature and pressure profiles and aerosol extinction is determined from the Raman signal which is less influenced by ozone under the assumption of an Ångström exponent for the wavelength dependence.

### 9.5.2 Measurement of Liquid Water

Raman signals from liquid water droplets in clouds have been first obtained as an enhanced water-vapor Raman signal corresponding to more than 100% relative humidity [34]. Because the measurements were done with a XeF excimer laser, which has several emission lines in the 348-to-353-nm region, and with relatively broad interference filters of 7–8 nm bandwidth, a considerable part of the liquid-water Raman band shown in Fig. 9.2 was detected in the water-vapor channel and led to an increased signal within the cloud. Demoz et al. [95] used the enhanced water-vapor Raman signal for the determination of cloud base height during light rain and cloud virga conditions, when other methods based on elastic backscattering gave questionable results. Whiteman and Melfi [35] suggested using the Raman signal from liquid water together with the elastic backscatter signal to determine liquid-water content, droplet radius, and droplet number density at the base of tropospheric clouds (the penetration depth is typically of the order of 100–200 m for optically dense water clouds).

For the determination of the liquid-water content from the Raman signal several requirements must be fulfilled. First of all, there must be proportionality between the Raman signal intensity and the liquid-water content. For single water droplets this condition is not met, since the Raman scattering cross section shows a variety of resonances with properties that depend on droplet size [96, 97]. However, the size distribution of cloud droplets is normally broad, and by integration over a range of sizes the cross section is found to be proportional to the droplet



volume [35, 97]. Second, an appropriate separation of the liquid-water Raman signal from the signal of water vapor is necessary. Because the two spectra overlap, the measurements must be carried out with high spectral resolution. Arshinov et al. [36] used a seeded Nd:YAG laser and a 32-channel receiving system to resolve the Raman spectrum between 3000 and 4200  $\text{cm}^{-1}$  frequency shift. A double-grating monochromator together with a holographic notch filter allowed a suppression of elastic scattering by 14 orders of magnitude. The photocathode of a 32-anode Hamamatsu photomultiplier was placed in the imaging plane of the spectrometer. In this way, spectrally resolved lidar signals could be measured in 32 channels simultaneously, and water-vapor and liquid-water signals could be separated.

The final point of interest is the sensitivity of the liquid-water backscatter spectrum to temperature. Whiteman et al. [98] showed that an isosbestic, i.e., a nearly temperature-insensitive, point in the liquid-water spectrum exists at a shift of 3425  $\text{cm}^{-1}$  (see Fig. 9.2). Thus, narrow-bandwidth detection of the liquid-water signal at that frequency will allow temperature-insensitive measurements.

The studies on the measurement of liquid water mentioned here represent an ongoing research. We can expect further results and new developments in the future.

## 9.6 Concluding Remarks

Nowadays, Raman lidar systems are instruments widely applied in atmospheric research with focus on water vapor, temperature (see Chapter 10), ozone, aerosols, and optically thin clouds (see Chapter 4). Because of their stable and robust setup, they are especially suitable for routine, automated, long-term atmospheric observations. The comparably small Raman scattering cross sections and the resulting low signal-to-noise ratios require appropriate temporal and spatial Raman signal averaging. The typical resolution of Raman measurements is 1–30 minutes in time and 50–300 m in space in the lower troposphere and 10 minutes to 2 h in time and 0.3–2 km in space in the upper troposphere and lower stratosphere. Raman lidar instruments are therefore used in climate and weather research to provide statistically significant information on the atmosphere, to establish climatologies of aerosols and water vapor [14, 15, 99], and to study mesoscale and large-scale processes such as frontal passages [100], hurricanes [101], and long-range

transport [16, 88, 102]. In the near range up to a few hundred meters distance and under a horizontal measurement geometry, water-vapor measurements for the study of turbulent fluxes were even made with a resolution of 1.5 m and 0.8 s [103].

Advanced Raman lidar systems are multichannel, multipurpose instruments which combine several Raman techniques and take measurements at several wavelengths [13, 16]. The characterization of the atmosphere in terms of the most important meteorological quantities, i.e., temperature and humidity, a comprehensive characterization of aerosol properties (see Chapter 4), and the investigation of dependencies between the observables, e.g., aerosol properties and relative humidity or ozone concentration and aerosol and cloud particle properties, are possible with such instruments.

A few pioneering steps in the application of Raman lidar systems in routine, long-term atmospheric monitoring may serve as a summarizing illustration of the potential of the technique:

- *Water vapor:* An operational Raman lidar has been working unattended and autonomously at the Cloud and Radiation Testbed site of the Atmospheric Radiation Measurement Program of the U.S. Department of Energy in Oklahoma since 1996. The lidar is one of several instruments in the program, the goal of which is to collect a 10-year data set on water vapor, aerosols, and clouds. The Raman lidar system is delivering the first long-term water-vapor climatology based on lidar measurements [11, 14, 99].
- *Ozone:* The Network for the Detection of Stratospheric Change, NDSC, applies lidar instruments to observe stratospheric ozone concentrations on a global scale. The Raman DIAL technique was implemented in these systems after the eruption of Mt. Pinatubo [104–106]. The network has meanwhile established an ozone climatology over more than a decade [107, 108].
- *Stratospheric Aerosol:* The aerosol load in the stratosphere and its decline after the eruption of Mt. Pinatubo in the Philippines in June 1991 had been observed with a Raman lidar for five years [48]. Aerosol extinction and backscatter profiles from Raman lidar measurements could be converted to particle effective radius, mass and surface-area concentrations [109] (see Chapter 4).
- *Tropospheric Aerosol:* The European Aerosol Research Lidar Network EARLINET carried out a three-year routine monitoring of the aerosol conditions over Europe. Out of the 21 stations of

the network, 11 use the Raman lidar technique to acquire aerosol extinction profiles. Several systems measure water vapor simultaneously. EARLINET is the first lidar network on a continental scale and the first network that applied Raman lidar as the basic monitoring technique [15].

The success of these applications will lead to the first Raman lidar system installed by a weather service for routine water-vapor and aerosol observations in the near future [110].

In addition to the well-established Raman lidar techniques mentioned above, there are more new and challenging applications under development. Recently, relative-humidity observations throughout the troposphere have been demonstrated by a combination of the water-vapor and temperature Raman techniques [16]. In conjunction with the spectrally resolved observation of liquid water with a 32-channel system, Arshinov et al. [36] found an unexpected return signal which they interpreted as scattering from water-molecule clusters, an intermediate state of water during transition between the gaseous and the liquid phase. Whiteman et al. [111] studied the feasibility of water-vapor Raman measurements from aircraft and found a reasonable performance. Together with the ongoing research on the measurement of liquid water (see Subsection 9.5.2) these examples may show some of the ways to go in the future for new applications of the lidar technique.

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# 10

## Temperature Measurements with Lidar

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### 10.1 Introduction

Temperature is a key parameter of the state of the atmosphere. Temperature data play an important role in such fields as atmospheric dynamics, climatology, meteorology, and chemistry, to name just a few. In addition to these direct geophysical applications, the atmospheric temperature profile is necessary as an input parameter to many remote-sensing techniques including lidar for the determination of other quantities. Examples are the measurement of the particle backscatter and particle extinction coefficients with Raman lidar [1], particle polarization [2], water-vapor mixing ratio with Raman lidar [3], or the measurement of trace-gas concentrations with differential-absorption lidar. Until now, temperature profiles of a model atmosphere or the results of radiosonde soundings (in the free troposphere and lower stratosphere) have usually been taken for this purpose. It is obvious that the quality of the results is considerably improved when the atmospheric temperature profile is measured at the same location during the same time interval.

### 10.2 Overview on Temperature Lidar Techniques

Today, lidar techniques for the remote sensing of atmospheric temperature profiles have reached the maturity necessary for routine observations. Stable and rugged systems have been employed successfully and advanced the understanding of atmospheric processes and climatology. At present, there are three lidar techniques available for



routine observations. Together, they cover a height range from the ground to the lower thermosphere: rotational Raman (for observations from the ground to the upper stratosphere), the integration technique (from the lower stratosphere up to the mesopause), and the resonance fluorescence technique (from the mesopause region to the lower thermosphere). As any optical measurement in the atmosphere that does not depend on the sun or some other celestial object as a radiation source, these techniques work best at nighttime when the background noise is low. However, high-power systems with spectrally narrow signal detection have to date been set up that also perform well under daytime conditions. Other lidar techniques for temperature profiling are under development such as high-spectral resolution lidar (HSRL) and differential absorption lidar (DIAL).

In this chapter, lidar techniques for temperature-profile measurements are reviewed. The reader will find a detailed description of integration lidar and rotational Raman lidar. The remaining techniques for lidar temperature profiling are covered in detail in other chapters of this book: the resonance fluorescence technique in Chapter 11, the HSRL technique in Chapter 5 and temperature measurements with DIAL in Chapter 8. An overview of the techniques presently available for temperature profiling with lidar is given in Table 10.1.

The integration technique uses a molecular backscatter signal. This signal can be either the Rayleigh band, the Cabannes line (sometimes also called “Rayleigh line”), a temperature-independent fraction of the pure rotational Raman band, a vibrational Raman band or line, or part of the central (“Gross”) Brillouin line. The intensity of a molecular lidar signal is proportional to the number density  $N$  of atmospheric molecules at height  $z$ ,  $N(z)$ . Under the assumption that the atmosphere is in hydrostatic equilibrium and with the initialization at a reference height, the temperature profile can be derived from  $N(z)$  and the ideal-gas law. To extend the height range of the integration technique downward below  $\sim 30$  km where particles are present in quantities sufficient to severely perturb Rayleigh integration lidar, an inelastic lidar such as the vibrational Raman signal of  $N_2$  can be used instead of the Rayleigh signal [4–6]. However, when aerosol extinction becomes significant compared with molecular extinction, this technique also fails.

Here rotational Raman (RR) lidar is the method of choice. With RR lidar, temperature measurements can be carried out not just in the clear atmosphere, but in aerosol layers and optically thin clouds as well. The technique is based on the fact that the intensities of the

**Table 10.1.** Overview of the lidar techniques for atmospheric temperature profiling. The height ranges are for the systems which were most advanced at the time of writing

| Technique                                                                    | Principle                                                                                                                                                                     | Comments                                                                                                                                                                                                   |
|------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Integration technique with elastic (Rayleigh or Cabannes) backscatter signal | <ul style="list-style-type: none"> <li>• Molecular lidar signal is proportional to molecular number density</li> <li>• Hydrostatic equilibrium is assumed</li> </ul>          | <ul style="list-style-type: none"> <li>• For measurements between <math>\sim 30</math> and <math>\sim 100</math> km</li> <li>• Observed height region must be free from particles</li> </ul>               |
| Integration technique with inelastic (Raman or Brillouin) backscatter signal | Same as above                                                                                                                                                                 | <ul style="list-style-type: none"> <li>• For measurements in the stratosphere</li> <li>• Can handle particle backscattering but no significant particle extinction</li> </ul>                              |
| Rotational Raman technique                                                   | <ul style="list-style-type: none"> <li>• Temperature dependence of the intensities of rotational Raman lines</li> <li>• Ratio of two signals</li> </ul>                       | <ul style="list-style-type: none"> <li>• For measurements in the troposphere and stratosphere</li> <li>• Measurements in clouds possible (details see text)</li> </ul>                                     |
| Brillouin–Doppler technique (HSRL ratio)                                     | <ul style="list-style-type: none"> <li>• Temperature dependence of the Doppler broadening of the Brillouin band</li> <li>• Ratio of two signals</li> </ul>                    | <ul style="list-style-type: none"> <li>• For measurements in the troposphere</li> <li>• Measurements in thin clouds possible</li> </ul>                                                                    |
| DIAL                                                                         | <ul style="list-style-type: none"> <li>• Temperature dependence of the strength of molecular absorption lines</li> </ul>                                                      | <ul style="list-style-type: none"> <li>• Feasibility demonstrated, first experimental data in boundary layer available</li> <li>• Difficulties in handling gradients of particle backscattering</li> </ul> |
| Resonance fluorescence                                                       | <ul style="list-style-type: none"> <li>• Temperature dependence of the Doppler broadening of resonance fluorescence of metal atoms</li> <li>• Ratio of two signals</li> </ul> | <ul style="list-style-type: none"> <li>• High-resolution measurements in heights where metal atoms are present with high mixing ratio, i.e., <math>\sim 75</math> km to 120 km</li> </ul>                  |

HSRL: High spectral resolution lidar (note that the integration technique with the use of a Brillouin signal is also called HSRL in the literature and must be distinguished from the HSRL ratio technique); DIAL: differential absorption lidar.

lines within the pure rotational Raman band exhibit different dependencies on temperature. The intensities of lines close to the incident laser line decrease with rising temperature, while the intensities of lines with large wavelength differences increase. This feature is a result of the population of rotational energy levels which is described by a

Maxwell–Boltzmann distribution. With the ratio of two RR lidar signals of opposite temperature dependence, one has a quantity with a temperature dependence that is independent of atmospheric transmission and range. In contrast to the integration technique, the RR technique does not use the assumption that the atmosphere is in hydrostatic equilibrium. Thus, even in turbulent layers no systematic measurement errors occur. In addition, as inelastic lidar signals, i.e., signals at wavelengths different from the laser wavelength are used, the rotational Raman technique can also be applied in heights in which particles are present because the backscatter signal of particles has the same wavelength as the laser. However, for RR lidar measurements in clouds to produce reliable results, the transmission of elastic-backscatter light in the rotational Raman channels must be very low ( $<10^{-7}$ ). As the rotational Raman line signals are only a few nm apart from the laser wavelength, the realization of such a system was quite an instrumental challenge. The problem is solved today using state-of-the-art optical components in the receiver.

The Brillouin–Doppler technique (see Chapter 5), also called “Rayleigh–Doppler technique” or “high-spectral-resolution lidar (HSRL) ratio technique,” uses the temperature dependence of the width, i.e., Doppler broadening, of the Cabannes line. Meanwhile, temperature measurements up to the lower stratosphere can be made with this technique using iodine vapor absorption cells of different temperatures to reject the elastic particle signal and to obtain two molecular lidar signals of different dependence on atmospheric temperature [7].

Also, differential absorption lidar (DIAL, see Chapter 8) can be used for temperature measurements. The temperature-dependent absorption of  $O_2$  lines in the near infrared is used for this purpose. Simulations, however, reveal that it is difficult to deal with the effect of Doppler-broadening of the Rayleigh backscatter signals with sufficient accuracy [8].

Finally, lidar temperature measurements around the mesopause and in the lower thermosphere are based on the Doppler-broadened lineshape of resonance fluorescence from atoms of metals such as Na, Ca, K, Fe which are present in varying concentrations between  $\sim 75$  and 120 km height (see Chapter 11). The large backscatter cross section of resonance fluorescence and the absence of particle scattering at these altitudes make the technique highly reliable. Daytime operation is possible by using, e.g., Faraday cells in the receiver which act as ultra-narrowband filters to exclude the sunlight and only transmit the lidar return signals.

## 10.3 The Integration Lidar Technique

### 10.3.1 Theory

A profile of the spatial density  $N(z)$  of atmospheric molecules can be derived from the elastic lidar return signal  $S(z)$  by rearranging the lidar equation, yielding

$$N(z) = \frac{C S(z) (z - z_0)^2}{\tau(z_0, z)^2}. \quad (10.1)$$

Here  $z_0$  is the altitude of the lidar above sea level.  $C$  includes all atmospheric and instrumental parameters that are constant with height during the time of the measurement.  $C$  also includes the extinction of the lidar signal in as far as it is due to particles and clouds; when defining  $C$  in this way, it is tacitly assumed that the integration from which the method borrows its name (see below) is carried out at elevations well above that part of the atmosphere in which clouds and aerosols are present.  $\tau(z_0, z)^2$  is the atmospheric roundtrip molecular transmission between the lidar and the measurement height  $z$ .  $C$  depends on the lidar system (laser power, receiver telescope area, receiver efficiency), on the type of signal detected (Rayleigh, Cabannes, Raman), the height resolution of the measured data, and the particle load of the atmosphere. As  $C$  is unknown and difficult to determine accurately enough, only a relative density profile can be deduced from the lidar data alone. To obtain absolute density values,  $C$  is quantified by normalizing  $N(z)$  either to a climatological model atmosphere or to radiosonde data at a reference height  $z_{\text{ref},1}$  that must be higher than all particle-containing height regions:

$$N(z) = \frac{N(z_{\text{ref},1}) S(z) (z - z_0)}{S(z_{\text{ref},1}) \tau(z_{\text{ref},1}, z)^2 (z_{\text{ref},1} - z_0)^2}, \quad (10.2)$$

where  $\tau(z_{\text{ref},1}, z)$  is the atmospheric transmission between  $z_{\text{ref},1}$  and  $z$ . However, as will be explained below, temperature measurements with the integration technique do not require the density profile itself but only the density ratio in two successive range bins.

If a Raman lidar signal is used for the temperature measurement, the difference in wavelength is accounted for by using  $\tau(\lambda_0, z_0, z) \tau(\lambda', z_0, z)$ , with  $\lambda_0$  for the laser wavelength and  $\lambda'$  for the wavelength of the Raman signal, instead of  $\tau(\lambda_0, z_0, z)^2$  or, as had been written for simplicity in Eqs. (10.1) and (10.2),  $\tau(z_0, z)^2$ . For  $\tau(\lambda_0, z_0, z)$  usually

one factor for the extinction of the signal due to Rayleigh scattering,

$$\tau_{\text{Ray}}(\lambda, z_0, z) = \exp \left( -\sigma_{\text{Ray}}(\lambda) \int_{z_0}^z N(\zeta) d\zeta \right), \quad (10.3)$$

and one factor for ozone absorption,

$$\tau_{\text{ozone}}(\lambda, z_0, z) = \exp \left( -\sigma_{\text{ozone}}(\lambda) \int_{z_0}^z N_{\text{ozone}}(\zeta) d\zeta \right), \quad (10.4)$$

are sufficient, since the laser wavelength used for integration lidar is normally not absorbed by other atmospheric molecules.  $N_{\text{ozone}}(z)$  can be taken, e.g., from radiosoundings, lidar measurements, or a model atmosphere. When inserting Eqs. (10.3) and (10.4) into Eq. (10.2), the resulting equation cannot be solved analytically for  $N(z)$ , thus the atmospheric density in the transmission term of Eq. (10.3) is as a rule approximated with data from a climatological model atmosphere. This procedure does normally not cause large errors. It is also possible to minimize the influence of model data at this point of the algorithm by an iterative approach: using the resulting  $N(z)$  of a first calculation as the input for the next yields a better second approximation, etc.

The temperature profile is derived from  $N(z)$  by making use of the ideal-gas law

$$p(z) = k N(z) T(z) \quad (10.5)$$

with the pressure  $p(z)$ , Boltzmann constant  $k$ , and temperature  $T(z)$  and with the barometric height formula

$$dp(z) = -\rho(z) g(z) dz \quad (10.6)$$

which is valid for the atmosphere under the condition of hydrostatic equilibrium.

$$\rho(z) = N(z) M \quad (10.7)$$

is the atmospheric density with  $M$  as the average molecular mass of the atmospheric constituents. The mixing ratio of the major atmospheric constituents  $\text{N}_2$ ,  $\text{O}_2$ , and Ar can be considered as constant at least up to the mesopause. Above, a small correction for the decrease of  $M$  with height is recommended. Equations (10.5)–(10.7) result in

$$T(z) = \frac{N(z_{\text{ref},2})}{N(z)} T(z_{\text{ref},2}) + \frac{M}{kN(z)} \int_{z_{\text{ref},2}}^z g(\zeta) N(\zeta) d\zeta. \quad (10.8)$$

Thus, with the input of  $N(z_{\text{ref},2})$  and  $T(z_{\text{ref},2})$ , the temperature profile  $T(z)$  can be derived in successive steps starting at the reference height  $z_{\text{ref},2}$ :

$$T(z_{i+1}) = \frac{N(z_i)}{N(z_{i+1})} T(z_i) + \frac{M}{kN(z_{i+1})} \bar{g} \bar{N} \Delta z. \quad (10.9)$$

Here  $z_i, z_{i+1}$  are the center heights of successive range bins of the lidar data,

$$\Delta z = z_{i+1} - z_i, \quad (10.10)$$

and  $\bar{g}$  and  $\bar{N}$  are the acceleration of gravity and molecular density, respectively, averaged between  $z_i$  and  $z_{i+1}$ .

For  $\bar{g}$  the approximation

$$\bar{g} = g(z_i + z_{i+1})/2 \quad (10.11)$$

is appropriate, whereas for  $\bar{N}$  the exponential dependence of  $N(z)$  with height must be taken into account, a linear fit would result in significant measurement errors. Using the approximation

$$N(z) = N(z_i) \exp(-b(z - z_i)) \Rightarrow N(z_{i+1}) = N(z_i) \exp(-b\Delta z), \quad (10.12)$$

where  $b$  is a constant, yields

$$\bar{N} = \int_{z_i}^{z_{i+1}} \frac{N(\zeta)}{\Delta z} d\zeta = \frac{N(z_i) - N(z_{i+1})}{b\Delta z}. \quad (10.13)$$

Since from Eq. (10.12) we have

$$\ln(N(z_{i+1})/N(z_i)) = -b\Delta z, \quad (10.14)$$

we get

$$\bar{N} = \frac{N(z_i) - N(z_{i+1})}{\ln(N(z_i)/N(z_{i+1}))}. \quad (10.15)$$

Thus, Eq. (10.9) can finally be written as

$$T(z_{i+1}) = \frac{N(z_i)}{N(z_{i+1})} T(z_i) + \frac{Mg((z_i + z_{i+1})/2)}{k} \Delta z \frac{(N(z_i)/N(z_{i+1})) - 1}{\ln(N(z_i)/N(z_{i+1}))}. \quad (10.16)$$

It is noteworthy that only the relative value of  $N(z)$  is used in Eq. (10.16) and  $T(z)$  is therefore independent of the normalization of the density profile at  $z_{\text{ref},1}$  in a first-order approximation, i.e., when a model atmosphere is used to calculate the Rayleigh extinction of the signal in Eq. (10.3).

In principle, the reference altitude  $z_{\text{ref},2}$  can be chosen at the upper or at the lower boundary of the altitude range in which the integration-technique measurements are made. The data are then derived successively downward or upward, respectively. However,  $T(z_{\text{ref},2})N(z_{\text{ref},2})$  must be highly accurate if profiles are to be generated upward because errors build up exponentially. On the other hand, when an upper reference height is chosen and the data are derived downward, errors in  $T(z_{\text{ref},2})N(z_{\text{ref},2})$  become quickly smaller. Due to this,  $T(z_{\text{ref},2})$  and  $N(z_{\text{ref},2})$  are usually taken at the upper boundary of the lidar data, e.g., from a climatological model atmosphere or from collocated measurements with other instruments like resonance fluorescence lidar or airglow imagers.

Errors which are caused by the initialization with  $T(z_{\text{ref},2})N(z_{\text{ref},2})$  were investigated, e.g., by Leblanc et al. [9] for downward integration. The authors used simulated data which were 15 K above the model atmosphere for all heights and found that the 15-K initialization error at 90 km decreased to 4 K at 80 km and to 1 K at 70 km. For real measurements the reference-height value should be much closer to the correct data than in this worst-case scenario so the actual downward-integration errors should be considerably smaller. It must be noted in this context that, when altitude-dependent signal-induced noise is present in the signals, inaccurate background subtraction can cause large errors. Signal-induced noise, when present, must be identified, and the data must be corrected.

The pressure profile  $p(z)$  is related to  $N(z)$  and  $T(z)$  via Eq. (10.5). Other formalisms which derive first a pressure profile and then the temperature profile [10] are equivalent to the algorithm discussed here.

### 10.3.2 Applications

Atmospheric temperature profiling with the lidar integration technique started in pre-laser times. As early as 1953, Elterman utilized a searchlight to obtain molecular density profiles. Via initialization with radiosonde data and the use of “physically acceptable” lapse rates,

he obtained temperature profiles up to 67.6 km [11, 12] and studied seasonal trends of temperature, density, and pressure [13]. First measurements with laser light were done in the 1960s with a Q-switched ruby laser by Sanford and reported in an early review paper [14]. At the beginning of the 1970s, pioneering integration-technique measurements with inelastic signals were carried out by Fiocco et al. [15]. These authors used a single-mode continuous-wave argon-ion laser to measure temperature and backscatter ratio in the troposphere. The particle backscattering contribution to the molecular signal was reduced with a Fabry–Perot interferometer. Later attempts to measure temperature profiles with Cabannes scattering and to block the particle backscatter signal with a Michelson interferometer did not succeed [16]. With the use of broadband dye lasers (which were originally developed for Na and Li density measurements in the mesosphere) Hauchecorne and Chanin obtained the first lidar temperature profiles between 35 and 70 km height [10] by detecting the Rayleigh backscatter signal from the middle atmosphere. In these altitudes there are virtually no particles present which at low heights perturb the measurements. Following this breakthrough, better-suited laser types like frequency-doubled Nd:YAG [17] and excimer lasers [18] were soon employed. In the following years, the high precision of these measurements which did not need to be calibrated and the ease of implementation of the technique on existing lidar systems started the study of a wide range of geophysical phenomena. These included gravity waves [19–22], stratospheric warming and planetary waves [23, 24], semi-diurnal and diurnal thermal tides [25–29], mesospheric inversions [30, 31], the oscillation of the 27-day solar cycle [32], the influence of the 11-year solar cycle [33, 34], climatology [35, 36], and long-term trends [37]. To ensure the quality of the measured data, sources of possible errors were investigated in detail [9, 38]. Rayleigh lidar systems serve today to evaluate and validate middle-atmosphere temperature measurements from satellites and are core instruments in the Network for the Detection of Stratospheric Change (NDSC).

## 10.4 Rotational Raman Lidar

### 10.4.1 Brief Historical Survey

The use of RR backscatter signals for atmospheric temperature profiling with lidar was originally proposed in 1972 by Cooney [39]. First



boundary-layer measurements were made with interference filters in the 1970s [40, 41] and later also with grating spectrometers [42]. With subsequent improvements in laser and receiver technology, the measurement range could be extended into the stratosphere [43, 44] and semi-automated, quite robust systems for tropospheric profiling were set up [45]. Advances in interference filter manufacturing allow today the design of receiver systems with very low losses in the separation of the lidar signals and massive reduction of the elastic backscatter crosstalk so measurements can be made even in clouds with a backscatter ratio of more than 50 without the need for corrections [46]. These systems can be built sufficiently rugged to work reliably under harsh conditions during field campaigns. The temporal resolution of today's most powerful systems is a few minutes within the troposphere at a height resolution of  $\sim 100$  m and a measurement uncertainty of  $\sim \pm 1$  K [47, 48]. The measurement range of these instruments extends up to the stratopause region for night-integrated observations. As an alternative to interference filters, double-grating spectrometers are also in use today which yield similar blocking power at slightly lower cost but also have lower receiver efficiency. A high-power interference-filter-based system can also measure in the daytime [47]. To improve the daytime performance, either a transmitter that emits at short wavelengths in the solar-blind spectral region [49] or a narrow-field-of-view receiver can be used. Furthermore, the spectral bandwidth of the receiver can be narrowed by separating the  $N_2$  RR lines by means of a Fabry–Perot interferometer (FPI) [50]. A first attempt using UV radiation from a narrow-band Raman-shifted KrF excimer laser and a thallium atomic vapor filter to block the elastic backscatter signal yielded measurements up to 1800 m height, but with large deviations of  $\pm 10$  K [51] from the data of a local radiosonde. The approach using an FPI succeeded in measurements throughout the troposphere with results very close to the data of a reference sonde [52]. The RR technique can conveniently be combined with other lidar techniques, thus allowing the setup of multiparameter Raman lidar systems. Standard laser sources with high emission power can be used with—as the only non-standard feature—frequency stabilization by injection seeding; this is very helpful for obtaining a stable calibration function. RR lidars have been used successfully for the investigation of a number of geophysical phenomena. One of the first and which is still in progress was the formation of polar stratospheric clouds in the arctic winter [53, 54]; here the unique capability of RR lidar to allow continuous monitoring of optical

particle properties and temperature in the same air masses proves highly beneficial.

### 10.4.2 Theory

In this section, a summary of the equations that describe the intensity of the atmospheric rotational Raman (RR) backscattering is given. The theory of Raman scattering is described in more detail in Chapter 9.

For simulation studies of RR temperature lidar, it is generally sufficient to consider air as a constant mixture of nitrogen and oxygen with relative volume abundances of 0.7808 and 0.2095, respectively. The remaining gases need not be considered. This also applies to water vapor. Its RR cross section is not much larger than the cross sections of  $N_2$  and  $O_2$  [55], but its relative amount is quite low even in conditions of water vapor saturation. Nitrogen and oxygen molecules can be treated here as simple linear molecules (SLM), i.e., linear molecules with no electronic momentum coupled to the scattering.

The rotational energy  $E_{\text{rot},i}(J)$  of a homonuclear diatomic molecule (like molecular nitrogen and oxygen) in the rotational quantum state  $J$  is [56]

$$E_{\text{rot},i}(J) = [B_{0,i}J(J+1) - D_{0,i}J^2(J+1)^2]hc, \quad J = 0, 1, 2, \dots, \quad (10.17)$$

where  $h$  is Planck's constant,  $c$  is the velocity of light, and  $i$  denotes the atmospheric constituent.  $B_{0,i}$  and  $D_{0,i}$  are the rotational constant and centrifugal distortion constant for the ground state vibrational level. On the frequency scale, the shift of the rotational Raman lines is independent of the wavelength of the exciting light  $\lambda_0$ . For the Stokes branch it is given by

$$\Delta\nu_{\text{St},i}(J) = -B_{0,i}2(2J+3) + D_{0,i}[3(2J+3) + (2J+3)^3], \\ J = 0, 1, 2, \dots \quad (10.18)$$

and for the anti-Stokes branch by

$$\Delta\nu_{\text{ASt},i}(J) = B_{0,i}2(2J-1) - D_{0,i}[3(2J-1) + (2J-1)^3] \quad \text{with} \\ J = 2, 3, 4, \dots \quad (10.19)$$

The differential backscatter cross section for single lines of the pure rotational Raman spectrum can be written as [57]

$$\left(\frac{d\sigma}{d\Omega}\right)_{\pi}^{\text{RR},i}(J) = \frac{112\pi^4}{15} \frac{g_i(J) h c B_{0,i}(\nu_0 + \Delta\nu_i(J))^4 \gamma_i^2}{(2I_i + 1)^2 k T} \times X(J) \exp\left(-\frac{E_{\text{rot},i}(J)}{kT}\right) \quad (10.20)$$

where for the Stokes branch

$$X(J) = \frac{(J+1)(J+2)}{2J+3} \quad \text{with } J = 0, 1, 2, \dots \quad (10.21)$$

and for the anti-Stokes branch

$$X(J) = \frac{J(J-1)}{2J-1} \quad \text{with } J = 2, 3, 4, \dots \quad (10.22)$$

$g_i(J)$  is the statistical weight factor which depends on the nuclear spin  $I_i$ .  $\nu_0$  denotes the frequency of the incident light,  $\gamma_i$  is the anisotropy of the molecular polarizability tensor,  $k$  is Boltzmann's constant and  $T$  is temperature. Equation (10.20) is valid if both polarization components of the RR signals are detected with the same efficiency in the lidar receiver. The depolarization of the pure rotational Raman wings is 3/4 [55], i.e., for linearly polarized incident light, a fraction of 4/7 of the backscatter signal intensity is polarized parallel and 3/7 perpendicular to the polarization plane of the incident light. To provide high signal intensities, RR lidars are usually designed to detect both polarization components with high efficiency.

The number of photons detected in an RR channel can be calculated with

$$S_{\text{RR}}(z) = S_0 \varepsilon \frac{A O(z)}{(z - z_0)^2} \Delta z N(z) \left[ \sum_{i=\text{O}_2, \text{N}_2} \sum_{J_i} \tau_{\text{RR}}(J_i) \eta_i \left(\frac{d\sigma}{d\Omega}\right)_{\pi}^{\text{RR},i}(J_i) \right] \times \tau_{\text{atm}}(z_0, z)^2 \quad (10.23)$$

where  $S_0$  is the number of transmitted photons,  $\varepsilon$  is the detector efficiency,  $A$  is the free telescope area,  $O(z)$  describes the overlap between transmitted laser beam and telescope,  $\Delta z$  is the height resolution,  $N(z)$  is the spatial density of air molecules,  $\tau_{\text{RR}}(J_i)$  is the transmission of the receiver at the wavelength of the RR line  $J_i$ ,  $\eta_i$  is the relative

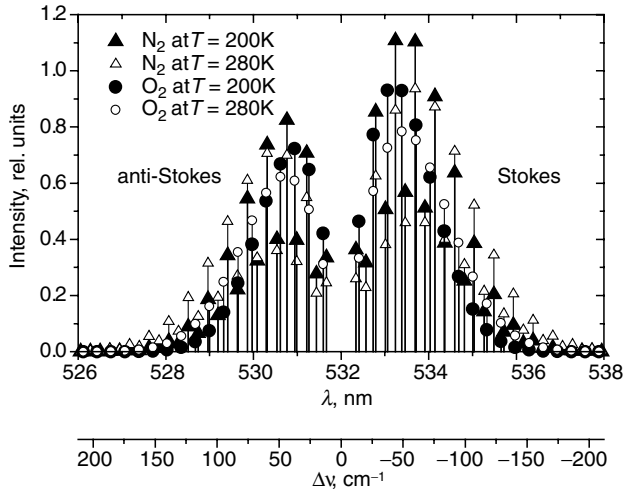


**Fig. 10.1.** Pure rotational Raman spectrum of air calculated for temperatures of  $T = 200\text{ K}$  and  $T = 280\text{ K}$  (intensity in relative units). The wavelength scale is for a laser wavelength of  $532\text{ nm}$ .

volume abundance of  $\text{N}_2$  and  $\text{O}_2$ , respectively, and  $\tau_{\text{atm}}(z_0, z)^2$  is the atmospheric roundtrip transmission.

Figure 10.1 shows the pure rotational Raman spectrum of air calculated for two temperatures, 200 and 280 K. The values employed for  $B_{0,i}$ ,  $D_{0,i}$ ,  $g_i(J)$ ,  $I_i$ , and  $\gamma_i^2$  are listed in Table 10.2. For simplicity, the cgs units used by the authors are quoted. In Chapter 9 it is shown how these values can be transformed into the SI system.  $\gamma_i$  depends weakly on wavelength [ $\gamma_{\text{O}_2}(\lambda = 488\text{ nm})/\gamma_{\text{O}_2}(\lambda = 647.1\text{ nm}) = 1.17 \pm 11\%$ ;  $\gamma_{\text{N}_2}(\lambda = 488\text{ nm})/\gamma_{\text{N}_2}(\lambda = 647.1\text{ nm}) = 1.13 \pm 11\%$ ] and is considered as constant with temperature in this context [57].

**Table 10.2.** Ground-state rotational and centrifugal distortion constants  $B_{0,i}$  and  $D_{0,i}$ , statistical weight factors  $g_i(J)$ , nuclear spin  $I_i$ , and the square of the anisotropy of the molecular polarizability tensor  $\gamma_i^2$ .

| Molecule     | $B_{0,i}$            | $D_{0,i}$                  | $g_i(J)$ |         | $I_i$ | $\gamma_i^2$                |
|--------------|----------------------|----------------------------|----------|---------|-------|-----------------------------|
|              | ( $\text{cm}^{-1}$ ) | ( $\text{cm}^{-1}$ )       | $J$ even | $J$ odd |       | ( $\text{cm}^6$ )           |
| $\text{N}_2$ | 1.98957 [60]         | $5.76 \times 10^{-6}$ [60] | 6        | 3       | 1     | $0.51 \times 10^{-48}$ [61] |
| $\text{O}_2$ | 1.43768 [56]         | $4.85 \times 10^{-6}$ [56] | 0        | 1       | 0     | $1.27 \times 10^{-48}$ [61] |

For RR temperature lidar the splitting of the O<sub>2</sub> RR lines in triplets [58] can be neglected because the two satellite lines located close to the main lines, at distances of 0.056 nm on either side, have intensities of only about 3% of the intensity of each central line. Pressure broadening of the RR lines [43] can also be disregarded when the transmission bands of the filters are broad compared to the width of the lines, which is  $\sim 2$  GHz. (With a suitable narrow-band receiver, however, one could also make use of the pressure broadening and measure temperature and pressure simultaneously [59].)

For atmospheric temperature profiling with the rotational Raman technique, usually the ratio

$$Q(T, z) = \frac{S_{RR2}(T, z)}{S_{RR1}(T, z)} \quad (10.24)$$

is used with  $S_{RR1}$  and  $S_{RR2}$  for two pure-rotational Raman signals of opposite temperature dependence. (One can also combine the signals in other ways which, however, gives no advantages.) In the ratio, altitude-dependent factors of the lidar equation cancel. For this it is assumed that the overlap functions are the same. For well aligned systems, this is the case above a certain minimum height. It is also assumed that atmospheric extinction of both signals is the same. Then the temperature dependence of the ratio  $Q$  is given by

$$Q(T) = \frac{\sum_{i=O_2, N_2} \sum_{J_i} \tau_{RR2}(J_i) \eta_i \left( \frac{d\sigma}{d\Omega} \right)_{\pi}^{RR,i}(J_i)}{\sum_{i=O_2, N_2} \sum_{J_i} \tau_{RR1}(J_i) \eta_i \left( \frac{d\sigma}{d\Omega} \right)_{\pi}^{RR,i}(J_i)} \quad (10.25)$$

where the formula for the Stokes or anti-Stokes differential backscatter coefficients have to be inserted depending on which branch of the pure RR spectrum is used.  $\tau_{RR1}(J_i)$  and  $\tau_{RR2}(J_i)$  are the transmissions of the lidar receiver at the wavelength of the rotational Raman line  $J_i$ . Figure 10.2 shows typical temperature dependencies of the RR signals used for atmospheric temperature measurements.

In order to provide the atmospheric temperature profile,  $Q(T)$  must be calibrated. The calibration can be done by characterizing the system parameters and using Eq. (10.25). This approach, however, may yield uncertainties on the order of a few K [44]. In practice, RR temperature lidar is therefore calibrated by comparison with data measured with other instruments such as local radiosondes; the temperature data of today's

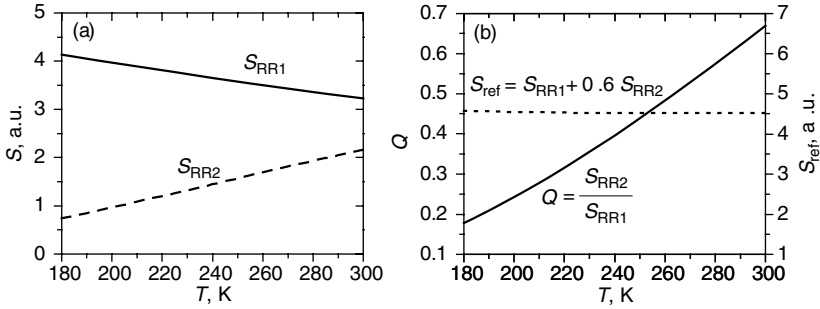


**Fig. 10.2.** (a) Typical intensities of the two pure-rotational Raman signals  $S_{RR1}$  and  $S_{RR2}$  as a function of temperature  $T$  [48]. (b) Signal ratio  $Q$  from which the atmospheric temperature is derived.  $S_{ref}$  can be used as a temperature-independent Raman reference signal for measuring extinction and backscatter coefficients of aerosols and cloud particles [47].

state-of-the-art radiosondes are accurate within tenths of a K provided the radiosonde itself has been accurately calibrated. Of course, the reference data used for the calibration should be taken as close in space and time as possible to the atmospheric column sensed by the lidar. How often an RR lidar systems needs to be recalibrated depends on the individual system. Provided that rugged mounts are used and the alignment of the lidar is not changed intentionally, the calibration of today's state-of-art systems remains virtually unchanged and only long-term degradations of the optical components may require recalibrations on a longer time scale.

For systems that detect only one RR line in each of the two RR channels Eq. (10.25) takes the simple form

$$Q(T) = \exp(a - b/T), \quad (10.26)$$

where the parameters  $a$  and  $b$  are both positive if  $J(S_{RR2}) > J(S_{RR1})$ .  $b$  is simply the difference of the rotational Raman energies of the extracted lines divided by  $k$ , and  $a$  is the logarithm of the ratio of all factors except the exponential term in Eq. (10.20). It is straightforward to use Eq. (10.26) also for systems with several lines in each of the RR signals [42]. But the obvious inversion of Eq. (10.26) which gives

$$T = \frac{b}{a - \ln Q} \quad (10.27)$$

then turns out to yield significant measurement errors, well in excess of 1 K (cf. Fig. 10.3) when measurements are made over an extended

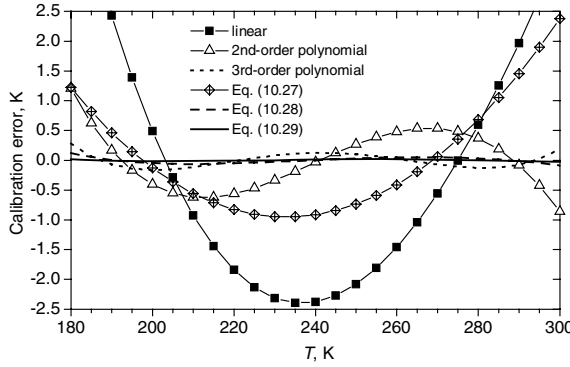


**Fig. 10.3.** Errors made with different calibration functions for rotational Raman temperature lidar.

range of temperatures. As the errors behave nearly as a second-order-polynomial function of temperature, it has been proposed to minimize calibration errors by a second calibration with such a second-order polynomial [43, 62], leading to a calibration function of the form

$$T = \frac{b}{a - \ln Q} + c \left( \frac{b}{a - \ln Q} \right)^2 + d \quad (10.28)$$

with the additional calibration constants  $c$  and  $d$ .

An even better calibration function, however, is found in the approach

$$Q = \exp \left( \frac{a'}{T^2} + \frac{b'}{T} + c' \right) \iff T = \frac{-2a'}{b' \pm \sqrt{b'^2 - 4a'(c' - \ln Q)}}, \quad (10.29)$$

which extends Eq. (10.26) to a second-order term in  $T$  and needs only three calibration constants  $a'$ ,  $b'$ ,  $c'$ . Fitting, as an example, the curve  $T(Q)$  shown in Fig. 10.2 with the different calibration functions, one gets the calibration errors shown in Fig. 10.3. The performances of polynomial calibration functions are also given for comparison. The single-line approach of Eq. (10.27) results here in errors of  $\sim \pm 1$  K for temperatures between 180 and 285 K, which is better than a linear calibration function. However, this relation is not generally valid [63]. For three calibration constants, Eq. (10.29) is superior to the second-order polynomial and even better than the third-order polynomial and the approach of Eq. (10.28), which both require four calibration constants. For Eq. (10.29), the temperature derived from the data with that

calibration function differs from the input temperature of the simulation by less than  $\pm 0.03$  K for temperatures between 180 and 300 K. With Eq. (10.28) or the third-order polynomial approach, the deviations are about  $\pm 0.10$  K and  $\pm 0.15$  K, respectively.

In conclusion, when data that cover a large range of temperatures ( $> 50$  K) are available for the calibration, Eq. (10.29) is the best-suited calibration of RR temperature lidar when several RR lines are sampled with each channel. Otherwise the single-line approach of Eq. (10.27) should be used to avoid large extrapolation errors.

### 10.4.3 Technological Considerations

#### Laser Wavelengths

Because it combines high output power with reliability and ease of handling, the Nd:YAG laser is at present the most widely used light source for Raman lidar systems. We theoretically investigate in the following the performance of RR temperature lidar with frequency-doubled and frequency-tripled radiation of a Nd:YAG laser, i.e., 355 nm and 532 nm. The fundamental emission at 1064 nm is less suitable because of low RR backscatter cross-sections and poorer detector efficiencies at this wavelength. Measurements with the fourth harmonic at 266 nm are limited to short range because light of this wavelength is strongly absorbed by ozone.

The ratio of photon counts in the rotational Raman channels is given by

$$\frac{S_{RR}(\lambda_1, J)}{S_{RR}(\lambda_2, J)} = \frac{d\sigma_J/d\Omega(\lambda_1)}{d\sigma_J/d\Omega(\lambda_2)} \left( \frac{\tau(\lambda_1)}{\tau(\lambda_2)} \right)^2 \frac{K(\lambda_1) S_0(\lambda_1)}{K(\lambda_2) S_0(\lambda_2)} \quad (10.30)$$

where  $S_{RR}(\lambda_i, J)$  are the numbers of photon counts detected and  $S_0(\lambda_i)$  are the numbers of photons transmitted by the lidar.  $J$  is the rotational quantum number,  $K(\lambda_i)$  denote the receiver efficiencies,  $(d\sigma_J/d\Omega)(\lambda_i)$  are the differential cross sections for pure rotational Raman backscattering, and  $\tau(\lambda_i)$  are the transmissions of the atmosphere with  $i = 1, 2$  and the primary wavelengths  $\lambda_1 = 532$  nm and  $\lambda_2 = 355$  nm. (The atmospheric transmission is approximately constant within the pure rotational Raman spectrum of one primary wavelength.)

The ratio of rotational Raman cross sections is

$$\frac{d\sigma_J}{d\Omega}(\lambda_1) \bigg/ \frac{d\sigma_J}{d\Omega}(\lambda_2) \approx (\lambda_2/\lambda_1)^4 = (2/3)^4 \approx 0.2. \quad (10.31)$$



Ozone absorption is  $<2\%$  for  $\lambda_1$  and  $<0.5\%$  for  $\lambda_2$  up to 30 km height [63]. So differences in ozone absorption can be neglected and the most important term that depends on height  $z$  for this comparison is the atmospheric transmission due to Rayleigh extinction. It is described by

$$\tau_{\text{Ray}}(z) = \exp \left( - \int_{z_0}^z N(\zeta) \sigma_{\text{Ray}}(\lambda) d\zeta \right), \quad (10.3)$$

where  $N(\zeta)$  is the molecular number density at height  $\zeta$ . It is taken here from a standard atmosphere. The total Rayleigh cross section can be calculated with

$$\sigma_{\text{Ray}}(\lambda) = \frac{8\pi^3(n(\lambda)^2 - 1)^2}{3\lambda^4 N_s^2} \left( \frac{6 + 3\gamma}{6 - 7\gamma} \right) \quad (10.32)$$

where  $\gamma = 0.0279$  is called “depolarization factor,”  $N_s = 2.547 \times 10^{25} \text{ m}^{-3}$  is the molecular number density, and  $n$  is the refraction index of air (both at standard conditions) with  $n(\lambda_1) - 1 = 2.78197 \times 10^{-4}$  and  $n(\lambda_2) - 1 = 2.85706 \times 10^{-4}$  [64, 65].

Typical values of the transmissions of the optical components in the receivers and typical detector efficiencies yield a ratio

$$\frac{K(\lambda_1)}{K(\lambda_2)} \approx 2. \quad (10.33)$$

For the same laser power, the ratio of the number of emitted photons is then given by

$$\frac{S_0(\lambda_1)}{S_0(\lambda_2)} = 1.5. \quad (10.34)$$

If laser pulse energies are the same for the two wavelengths, we thus have

$$\frac{P_{\text{RotRam}}(\lambda_1, J)}{P_{\text{RotRam}}(\lambda_2, J)} = 0.6 T^*(z) \quad (10.35)$$

with the height-dependent term

$$T^*(z) = (\tau(\lambda_1)/\tau(\lambda_2))^2 \quad (10.36)$$

which describes the effect of atmospheric transmission.

$T^*(z)$  and  $S_{\text{RR}}(\lambda_1, J)/S_{\text{RR}}(\lambda_2, J)$  as a function of height are shown in Fig. 10.4. Rotational Raman signals from altitudes above 20 km are  $\sim 60\%$  stronger if a primary wavelength of  $\lambda_1 = 532 \text{ nm}$  instead of  $\lambda_2 = 355 \text{ nm}$  is used. This means that it is advantageous to use 532 nm as the

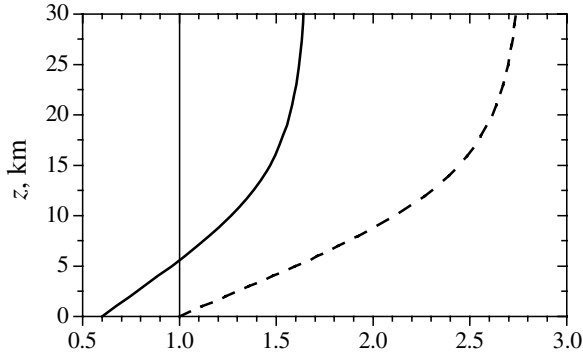


**Fig. 10.4.** Calculated ratio of rotational Raman signal intensities  $S_{RR}(\lambda_1, J)/S_{RR}(\lambda_2, J)$  (solid curve) and atmospheric transmission factor  $T^*(z)$  (dashed curve) versus height  $z$  with primary wavelengths  $\lambda_1 = 532$  nm and  $\lambda_2 = 355$  nm for equal laser power (see text for further details).

primary wavelength for a system aiming at high performance in the upper troposphere and in the stratosphere, whereas 355 nm is better suited for temperature measurements in the lower troposphere if the same laser power is available at both wavelengths. These results, however, were obtained under the assumption of Eq. (10.33). With future improvements in receiver technology, this assumption must certainly be reevaluated, which will extend the range for which UV primary wavelengths are superior for RR lidar. First measurements with a system not yet optimized were very promising and have already shown the general feasibility of using 355 nm as the primary wavelength for RR lidar [66].

In addition to signal intensity, it is also important to consider possible differences of the blocking of the elastic backscatter light in the RR channels. The molecular elastic backscatter signal is proportional to  $\lambda_0^{-4}$ , just like the pure rotational Raman signal. Thus no differences in relative signal intensity exist and the relative blocking required for merely the molecular elastic signal is independent of laser wavelength. For particle backscattering, the wavelength dependence varies with size, composition, and shape of the particles. One simplifying rule says that, if the average particle size is significantly larger than both laser wavelength (which is the case for most types of clouds and many types of aerosols), then the relative intensity of the particle signal is lower in the UV than in the visible. There is thus an advantage for a UV primary wavelength under the same atmospheric conditions. On the other hand, the blocking of the elastic backscatter signal in the RR channels that can be achieved today is also lower in the UV which tends to compensate that advantage.

## Required Suppression of the Elastically Backscattered Light in the Rotational Raman Channels

The major technical challenge for temperature measurements with the rotational Raman technique is to achieve sufficient blocking of the elastic backscatter signal in the rotational Raman channels. The problem is particularly serious in the low- $J$  channel because of its close proximity ( $<30\text{ cm}^{-1}$  or  $<1\text{ nm}$  at  $\lambda = 532\text{ nm}$ ). In this section we present a calculation which attenuation is needed. This point has risen considerable controversy in the literature [42, 67]. We show that to ensure measurement errors  $<1\text{ K}$  in clouds up to a backscatter ratio ( $R_{532}$ ) of 50, a blocking by at least seven orders of magnitude is required. This requirement can be met in practice with today's state-of-the-art optics (see Section 10.5).

The backscatter ratio  $R_{532}$  is defined as the ratio of total backscatter intensity to molecular backscatter intensity.\* The optical thickness  $\chi$  of the rotational Raman channels at the primary wavelength  $\lambda_0$  is

$$\chi = -\log_{10}(\tau(\lambda_0)) \quad (10.37)$$

where  $\tau(\lambda_0)$  is the filter transmission. For the calculation, we first simulate the pure rotational Raman signal of air for different temperatures. Then we multiply the spectra with the transmission curves of the receiver channels to get the intensities of the unperturbed rotational Raman signals. The results are used for deriving the calibration function. We add an offset to the unperturbed signals of a single input temperature according to the assumed leakage of the elastic signal. The “measured” temperature is then calculated with the perturbed signals via the calibration function. The leakage measurement error is the difference between the input temperature and the temperature which would be measured with the signals including elastic-signal leakage.

As an example, Fig. 10.5 shows the results of such a calculation for a backscatter ratio  $R_{532} = 50$  and input temperatures of 180 K and 250 K and the receiver parameters taken from Ref. 46. The leakage measurement error is zero if the ratio of elastic offset signals is the same

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\* The backscatter ratio, i.e., the ratio of total backscatter signal to molecular backscatter signal, is not consistently defined in the literature. Depending on the system in use, different receiver spectral widths result in the extraction of different fractions of the pure rotational Raman spectrum (PRRS) for the total backscatter signal. As the PRRS is about 3% of the total molecular backscatter signal the differences, however, are small.

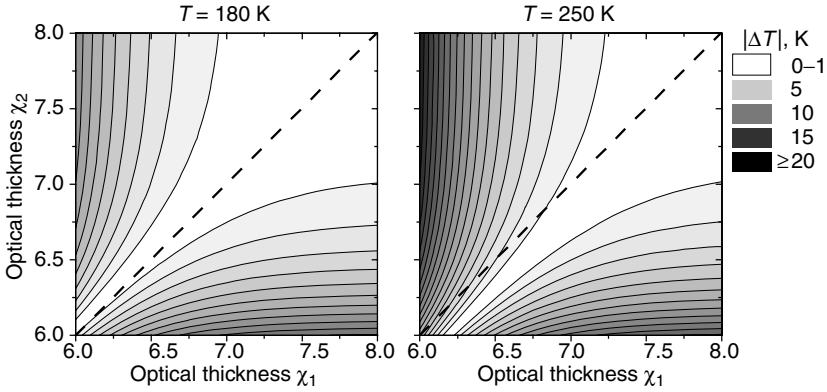


**Fig. 10.5.** Calculated leakage measurement error  $\Delta T$  for  $R_{532} = 50$  versus the primary-wavelength optical thicknesses  $\chi_1$  and  $\chi_2$  of the two rotational Raman channels for two “input” temperatures,  $T = 180$  K and  $T = 250$  K. Error contour lines are plotted at intervals of 1 K. Absolute values are given.  $\Delta T$  is negative in the upper left and positive in the lower right region of the plots. Dashed lines mark  $\chi_1 = \chi_2$ .

as the ratio of the unperturbed rotational Raman signals. This illustrates directly that the leakage error itself depends on temperature and on the parameters of the individual receiver. If the RR signal intensities are approximately the same within the range of atmospheric temperatures, similar values of elastic backscatter signal blocking are desirable. Ironically, a combination of a lower blocking in one and a higher blocking in the other channel may result in a larger leakage error than the same lower blocking value in both channels.

Figure 10.6 shows leakage errors for equal blocking  $\chi$  of the two RR channels for different values of “input” temperature. Even if the

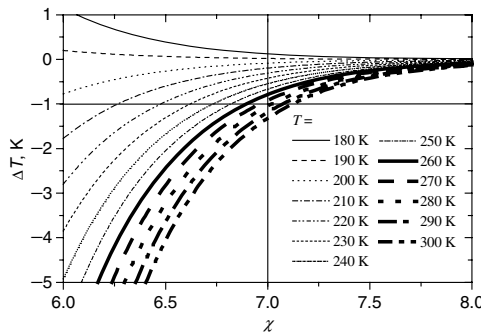


**Fig. 10.6.** Calculated leakage measurement error  $\Delta T$  for  $R_{532} = 50$  against optical thickness of the two rotational Raman channels  $\chi = \chi_1 = \chi_2$  at different temperatures  $T$ .

blocking for both RR channels is the same, the sign of the leakage errors is not necessarily the same for all temperatures. With the filter parameters used for this calculation, which may be considered as typical for RR lidar,  $|\Delta T|$  is  $< 1$  K between 180 and 280 K for  $R_{532} = 50$  with  $\chi \geq 7$  for both rotational Raman channels. In conclusion, if the elastic backscatter signal is blocked in the RR channels by at least seven orders of magnitude, leakage errors are small even in heights of significantly enhanced particle backscatter signals.

### Center Wavelengths and Bandwidths of the RR Channels

Next, we investigate the important question which filter center wavelengths ( $CWL_{RR1}$  and  $CWL_{RR2}$ ) and channel passbands (full-width-at-half-maximum bandwidths  $FWHM_{RR1}$  and  $FWHM_{RR2}$ ) yield the minimum statistical temperature error of RR temperature measurements. We can limit the calculations to combinations of the filter parameters with  $CWL_{RR1}$ ,  $CWL_{RR2}$  in regions of opposite temperature dependence and  $FWHM_{RR1}$ ,  $FWHM_{RR2}$  sufficiently small that  $\lambda_0$  is out of the filter transmission band.

The majority of today's RR lidar systems extract parts of only the anti-Stokes branch of the pure rotational Raman spectrum (PRRS) in order to avoid the risk of interference with aerosol fluorescence. In addition, when anti-Stokes signals are extracted and an interference-filter-based receiver is employed, the distance of the filter center wavelengths to the laser wavelength can be increased by increasing the angle of incidence onto the filters. This allows to fine-tune the transmission wavelength when the blocking at the laser wavelength must be increased.

The  $1\text{-}\sigma$  uncertainty of a photon counting signal  $S$  follows Poisson statistics and is therefore given by

$$\Delta S = \sqrt{S}, \quad (10.38)$$

which yields, for the uncertainty of the RR temperature measurement,

$$\begin{aligned} \Delta T &= \frac{\partial T}{\partial Q} Q \cdot \sqrt{\frac{1}{S_{RR1}} + \frac{1}{S_{RR2}}} \\ &= \left( \frac{\partial S_{RR1}}{\partial T} \frac{1}{S_{RR1}} - \frac{\partial S_{RR2}}{\partial T} \frac{1}{S_{RR2}} \right)^{-1} \sqrt{\frac{1}{S_{RR1}} + \frac{1}{S_{RR2}}}, \quad (10.39) \end{aligned}$$

where

$$Q = \frac{S_{RR2}}{S_{RR1}} \quad (10.24)$$

is the ratio of the two signals  $S_{RR1}$  and  $S_{RR2}$  of lower and higher RR quantum-number transition channels. We can approximate the derivatives in Eq. (10.39) with the corresponding differences such that

$$\frac{\partial T}{\partial Q} \approx \frac{T_2 - T_1}{Q(T_2) - Q(T_1)}. \quad (10.40)$$

It is appropriate to use a peak transmission  $\tau = 1$  and an out-of-band transmission  $\tau = 0$  in this simulation. With the use of, e.g., Gaussian-shaped filter transmission curves to account for the slope of the filter edges, the resulting data would just be smoother, which would neither improve the clarity of the results nor change the conclusions.

Figure 10.7 shows the temperature sensitivity

$$\frac{\partial S}{\partial T} \approx \frac{S(T_2) - S(T_1)}{T_2 - T_1}, \quad (10.41)$$

for different parts of the RR spectrum when  $T_1 = 235$  K and  $T_2 = 240$  K. For these temperatures, regions with negative sign of  $\partial S/\partial T$  are found at center wavelengths between  $\sim 530.5$  nm and  $\sim 534.5$  nm and positive

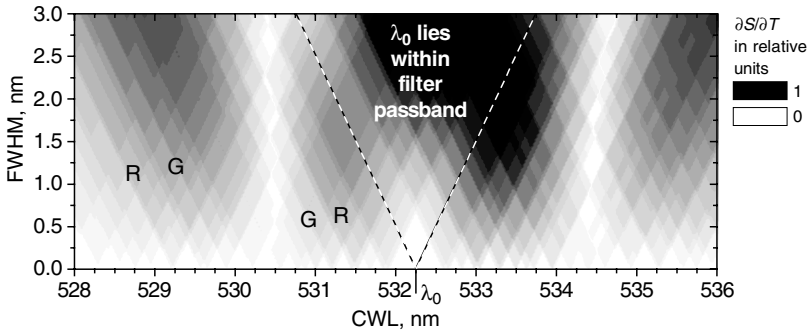


**Fig. 10.7.** Calculated temperature sensitivity  $\partial S/\partial T$  of atmospheric pure rotational Raman signals versus center wavelength CWL and bandpass full width at half maximum FWHM. The laser wavelength  $\lambda_0$  was set to 532.25 nm, temperatures to  $T_1 = 235$  K and  $T_2 = 240$  K. Calculation stepwidth was 0.025 nm. Absolute values only are plotted;  $\partial S/\partial T$  is negative for CWLs between  $\sim 530.5$  nm and  $\sim 534.5$  nm and positive elsewhere. The parameters of two interference-filter-based RR lidars are marked with ‘G’ [46] and ‘R’ [48], respectively.

$\partial S/\partial T$  elsewhere. For RR temperature measurements, one RR signal should be taken from each of these regions. Within the data range displayed in Fig. 10.3, the temperature sensitivity of the anti-Stokes signals is most pronounced around ( $\text{CWL}_{\text{RR1}} = 531.30 \text{ nm}$ ,  $\text{FWHM}_{\text{RR1}} = 1.2 \text{ nm}$ ) and ( $\text{CWL}_{\text{RR2}} = 529.30 \text{ nm}$ ,  $\text{FWHM}_{\text{RR2}} = 3.0 \text{ nm}$ ). However, while for the RR1 channel a maximum temperature sensitivity exists because of the boundary imposed by the laser wavelength, the RR2 filter has no such optimum because the extraction of more lines with higher rotational quantum number always improves the sensitivity for this channel. Clearly, it is not the temperature sensitivity alone that must be optimized, but also the intensity of the RR signals, as is described by Eq. (10.39); regions of the RR spectrum with highest signal intensity and highest temperature sensitivity are not identical.

As an example, Fig. 10.8 shows the dependence of the statistical temperature error on filter center wavelengths for filter bandwidths fixed at  $\text{FWHM}_{\text{RR1}} = 0.6 \text{ nm}$  and  $\text{FWHM}_{\text{RR2}} = 1.2 \text{ nm}$  and for  $T_1 = 235 \text{ K}$  and  $T_2 = 240 \text{ K}$ . These bandwidths were used by two systems [46, 48] which also meet the requirement of high blocking of the elastic

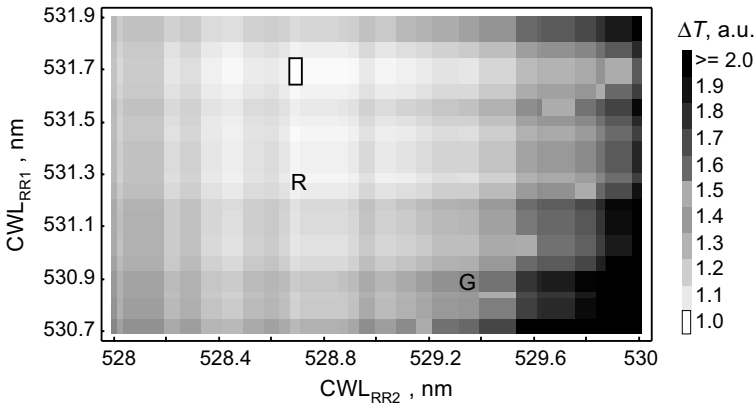


**Fig. 10.8.** Result of optimization calculations for the center wavelength (CWL) of both rotational Raman channels [48]: Statistical measurement uncertainty  $\Delta T$  versus filter center wavelengths  $\text{CWL}_{\text{RR1}}$  and  $\text{CWL}_{\text{RR2}}$  for temperatures  $T_1 = 235 \text{ K}$  and  $T_2 = 240 \text{ K}$ . For the calculation, the filter transmission curves were approximated by rectangular filter passbands with widths of 0.6 and 1.2 nm for the first and second rotational Raman channel, respectively. Calculation step width was 0.025 nm. Values are given relative to the minimum error near  $\text{CWL}_{\text{RR1}} = 531.7 \text{ nm}$ ,  $\text{CWL}_{\text{RR2}} = 528.7 \text{ nm}$  ( $\square$ ). ‘G’ [46] and ‘R’ [48] mark the CWLs of two RR lidar systems.

backscatter light discussed above. For these temperatures, the statistical temperature error becomes smallest around ( $CWL_{RR1} = 531.7$  nm,  $CWL_{RR2} = 528.7$  nm).

Optimum filter parameters depend on the temperature that is to be measured. Fortunately this dependence is not very strong within the range of atmospheric temperatures. For example, optimum CWLs for  $T_1 = 185$  K and  $T_2 = 190$  K, the condensation conditions of polar stratospheric clouds, are 531.70 and 529.35 nm. Using filters optimized for 240 K ( $CWL_{RR1} = 531.7$  nm,  $CWL_{RR2} = 528.7$  nm) increases measurement uncertainties at 185 K by a mere 10%.

### 10.5 Technical Implementation: Combined Lidar for Temperature Measurements with the Rotational Raman and the Integration Technique

Because of the high requirements on separation and blocking power of the filters which were only met in recent years, RR temperature lidars have entered the lidar scene at a time when other parameters were already obtained with lidar on a routine basis. It was thus most beneficial that some of these RR lidars could be designed as add-on systems to existing lidar facilities. Many components including the radiation source could thus be shared between the RR and the existing lidar. The system presented in this section as an example for a practical RR temperature lidar is such a combined instrument.

The lidar described in the following was designed for simultaneous temperature measurements with the RR technique and the integration technique at the Radio Science Center for Space and Atmosphere, Kyoto University, Japan [47, 48]. It is not the first instrument combining these techniques [63, 68, 69], but provides the most intense RR signals at the time when this text is written. It may illustrate here the state-of-the-art performance of such a combined system. The RASC lidar is located at the MU (Middle and Upper atmosphere) Radar Observatory at (34.8 °N, 136.1 °E) in Shigaraki, Japan. The receiver scheme for the rotational Raman channels follows a design developed at GKSS Research Center, Germany [46]. The use of narrow-band interference filters allows a consecutive setup of the elastic channel and the two rotational Raman channels. This scheme yields both high efficiency and low cross-talk effects.

A sketch of the RASC lidar is given in Fig. 10.9. The lidar transmitter is an injection-seeded Nd:YAG laser. The seeder wavelength is



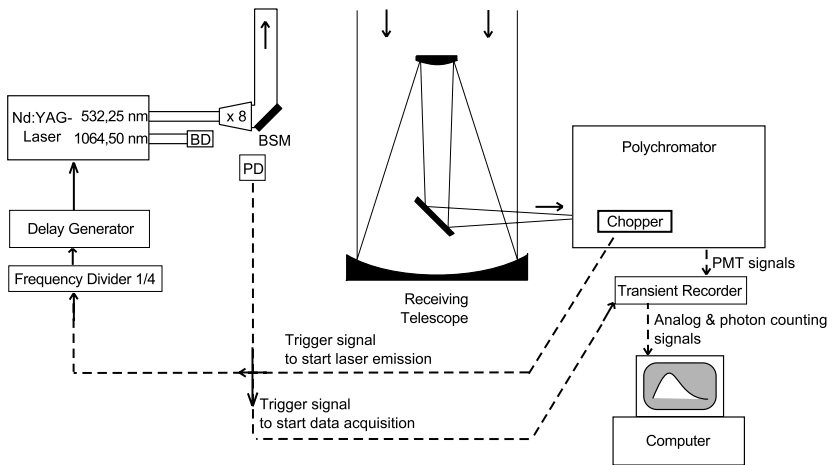


**Fig. 10.9.** Setup of the RASC lidar. BD beam dump, PD photodiode, BSM beam steering mirror, PMT photomultiplier tube [48].

1064.50 nm in vacuum yielding a wavelength of  $\lambda_0 = 532.25$  nm for the second-harmonic radiation which is used as the primary wavelength of the lidar. The laser repetition rate is 50 Hz with an output power of about 30 W at  $\lambda_0$ . Light backscattered from the atmosphere is collected with a Cassegrainian telescope with primary-mirror diameter of 0.82 m. A filter polychromator separates the signals which are finally detected with photomultipliers (PMTs). Data acquisition electronics store the data in both photon counting mode and analog mode with a temporal resolution of typically 3000 laser pulses (1 minute) and a range resolution of 72 m up to a height of 147 km above the lidar, the best possible resolutions in time and height being 10 s and 9 m with the data acquisition system used. The raw data are later smoothed with sliding-average algorithms that are adapted to the requirements of the current atmospheric investigation. The laser emission is synchronized to the mechanical chopper blade which is placed in the branch of the high-altitude elastic channel to protect the PMT from the intense low-altitude signals.

The separation of the elastic signal and the RR signals is done with multi-cavity interference filters (BS3, BS4a, BS4b, and BS5 in Fig. 10.10). The main parameters of the filters are given in Table 10.3. As the reflectivity of each component is near unity for the signals that follow in the line, signal separation is very efficient. In addition, by extracting the elastic signal upstream of the RR signals, the elastic signal intensity in the reflected beam is already attenuated by a factor of 9 when it

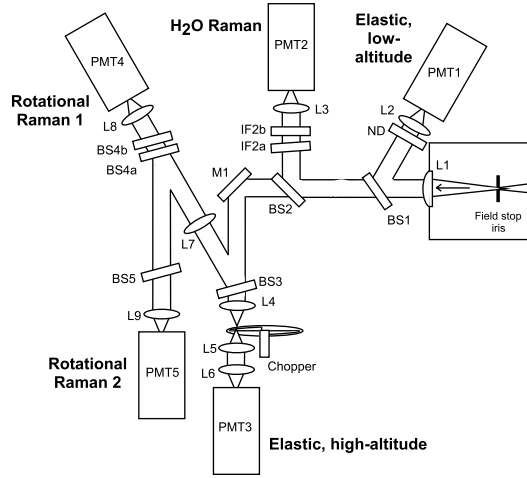


**Fig. 10.10.** Setup of the RASC lidar polychromator. L1–L9 lenses, IF1–IF2b interference filters, BS1–BS5 beamsplitters, ND neutral density attenuator, PMT1–PMT5 photomultiplier tubes for the signals indicated [48].

reaches the RR channel filters. This feature enhances the elastic-signal blocking for the RR channels and reduces the requirements for the filters used to extract the RR signals. A total receiver blocking of seven orders of magnitude, which was aimed at (see Subsection 10.4.3), is indeed reached. BS3 to BS5 are mounted at small angles of incidence of  $4.8^\circ$ ,  $5.0^\circ$ , and  $7.2^\circ$ . By changing these angles the center wavelengths of the extracted signals can be tuned, e.g., to optimize the blocking

**Table 10.3.** Properties of the beamsplitters BS3, BS4a, BS4b, and BS5 used in the receiver of the RASC lidar. The laser emission, peak transmission of BS4a and BS4b, and peak transmission of BS5 are at the wavelengths of 532.25 nm, 531.1 nm, and 528.5 nm, respectively. AOI angle of incidence, CWL center wavelength, FWHM full width at half maximum,  $\tau$  transmission,  $\rho$  reflectivity.

| Parameter                                    | Wavelength, nm | BS3                              | BS4a & BS4b combined | BS5                      |
|----------------------------------------------|----------------|----------------------------------|----------------------|--------------------------|
| AOI, degrees                                 |                | 4.8                              | 5.0                  | 7.2                      |
| CWL, nm                                      |                | 532.34                           | 531.14               | 528.76                   |
| FWHM, nm                                     |                | 0.80                             | 0.65                 | 1.10                     |
| Transmission/<br>reflection at<br>wavelength | 532.25         | $\tau = 0.82$ ,<br>$\rho = 0.11$ | $\tau < 10^{-6}$     | $\tau < 10^{-6}$         |
|                                              | 531.1          | $\rho > 0.95$                    | $\tau = 0.72$        | $\tau < 2 \cdot 10^{-4}$ |
|                                              | 528.5          | $\rho > 0.96$                    | $\rho > 0.96$        | $\tau = 0.87$            |
|                                              |                |                                  |                      |                          |

at the laser wavelength or to improve the receiver performance for measurements at varying temperature ranges of interest.

The elastic signal from low altitudes is too intense to be detected in photon counting mode with PMT 3. But that is no problem for low-altitude data alone: If the PMT is not protected from the intense low-altitude radiation, the signals also from higher altitudes are affected by signal-induced noise. Thus a chopper with 100 Hz rotation frequency is employed. The beam is focused on the outer edge of the blade opening at a distance of about 5 cm from the center to allow fast “switching.” As the elastic signal from low altitudes is blocked at PMT 3, it is detected in a separate branch with PMT 1. A glass plate (BS1) reflects a small fraction of the total signal intensity out of the beam. The two RR signals of different temperature-dependence are used for both atmospheric temperature measurements and to derive a temperature-independent Raman reference signal. This signal serves to measure the particle extinction coefficient and the particle backscatter coefficient with smaller errors that can be done with the common vibrational Raman technique [47]. The RASC lidar also detects the vibration–rotation Raman signal from water vapor for the measurement of the water vapor mixing ratio. Combining these data with the measured temperature profile yields relative humidity.

The intensities of the rotational Raman signals of the RASC lidar are shown in Fig. 10.11 for typical clear-sky conditions except for a thin cirrus. The RR signals are factors of  $\sim 2500$  (RR1) and  $\sim 5000$  (RR2) less intense than the elastic signal. Nevertheless, they allow RR lidar temperature measurements with high resolution both day and night. For a solar elevation angle of 45 degrees, e.g., the  $1\text{-}\sigma$  statistical uncertainty is 1.4 K at a height of 3 km for a measurement resolution of 300 m in 6 minutes with a receiver field-of-view of 1 mrad full-angle [47]). The high blocking of the strong elastic backscatter signal for the Raman channels allows also measurements in thin clouds. No cross-talk is seen up to a backscatter ratio of  $\sim 45$ . Even when cross-talk is found, this does not render the RR data useless. The amount of leakage can be quantified and the leaked-through elastic signals can be subtracted from the apparent RR channel signals [47].

### 10.5.1 State-of-the-Art Performance

A temperature profile measured with the RASC lidar is shown in Fig. 10.12. The height ranges of the RR data and the integration technique

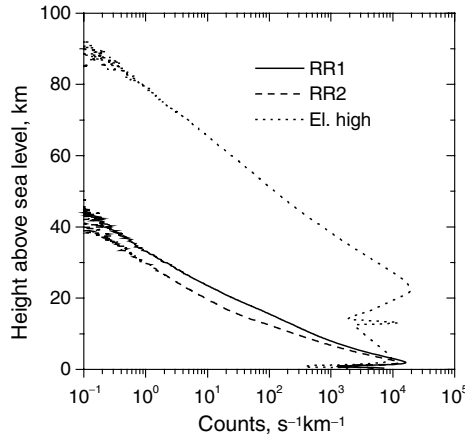


**Fig. 10.11.** Intensities of the RASC lidar signals for the temperature measurements: rotational Raman signals (RR1 and RR2) and high-altitude elastic signal (El. high). For this plot, 72 minutes (216,000 laser pulses) of nighttime lidar data were taken with a height resolution of 72 m, summed, the background was subtracted, and the data were finally smoothed with a sliding average of 360 m. The photon emission rate of the laser is  $\sim 8 \times 10^{19}$  photons/s. In the high-altitude elastic signal, the effect of the chopper can be seen below  $\sim 25$  km and the signature of a cirrus cloud in  $\sim 13$  km height.

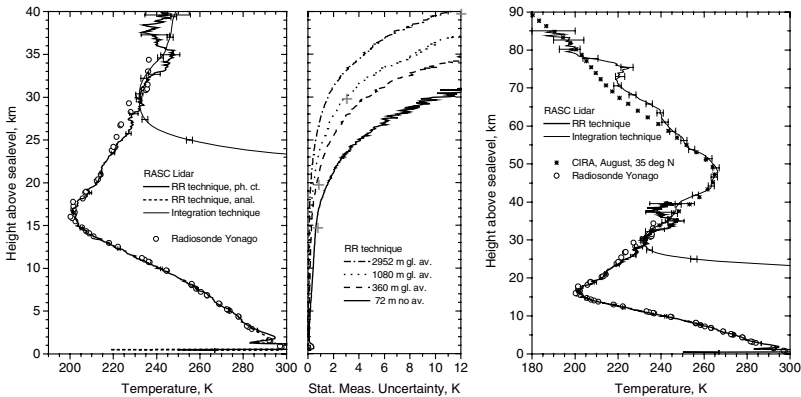


**Fig. 10.12.** Simultaneous temperature measurements with rotational Raman technique and with integration technique (signals see Fig. 10.11). Profiles of a climatological model atmosphere (CIRA-86 for  $35^\circ\text{N}$  and the month of the lidar measurements) and of a radiosonde are shown for comparison. Rotational Raman temperature data: height resolution of 72 m up to 15 km height, 360 m between 15 and 20 km height, 1080 m between 20 and 30 km height, and 2952 m above 30 km. Height resolution of the integration technique data is 2952 m. Error bars show the  $1-\sigma$  statistical uncertainty of the measurements [48].

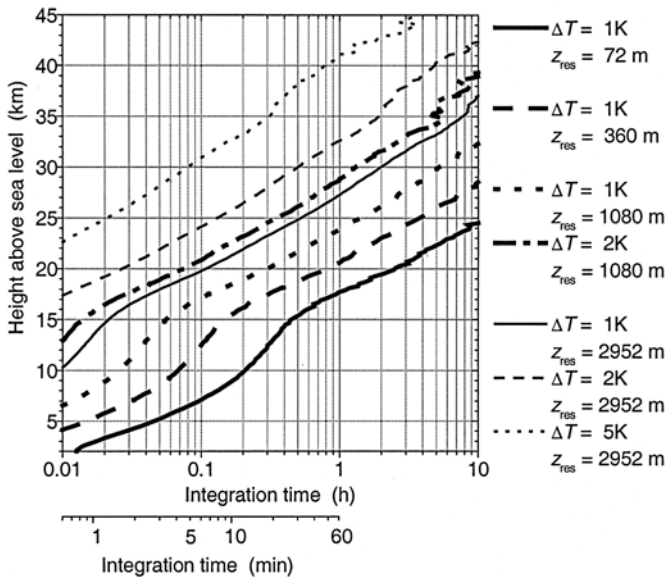


**Fig. 10.13.** Relation between integration time, height resolution, and statistical uncertainty of the temperature measurements with rotational Raman technique for the RASC lidar (calculated with the same data as those used for Fig. 10.11) [48].

data overlap well. In the overlap region, the results of the two techniques coincide within the statistical uncertainty. At heights below  $\sim 29$  km the chopper attenuates the signal and interferes with the integration-technique data. The RR temperature data below  $\sim 3$  km are affected by the nonlinearity of the photon counting electronics (although the signals were corrected for deadtime effects using the model of a nonparalizable detector); here the analog data remain reliable down to  $\sim 2$  km above sea level which is  $\sim 1.6$  km above the lidar. For lower altitudes, the overlap geometry between the laser beam and the field of view of the telescope is different for the two RR channels and causes deviations with this system. For measurements at low altitudes a coaxial arrangement of the outgoing laser beam with the receiving telescope would be the preferred geometry to allow measurements from close to the ground upward.

The relation between integration time, height resolution, and statistical uncertainty of the RR temperature measurements of the RASC lidar can be seen in Fig. 10.13. With, e.g., 5 minutes integration time and a

height resolution of 72 m, the  $1-\sigma$  statistical uncertainty of the data is  $<1$  K up to  $\sim 7$  km altitude. For measurements in the stratosphere, e.g., at 23 km altitude an integration time of 1 hour results in 1 K uncertainty for 1080 m averaging, while for 45 km altitude and 2952 m smoothing 4 hours of integration time yield 5 K uncertainty.

This example as well as many of the applications briefly mentioned in Subsection 10.4.1 show that rotational Raman lidar is probably the most accurate, most precise and cheapest method for the determination of vertical profiles of atmospheric temperature in the troposphere and lower stratosphere with or without the presence of aerosols and optically thin clouds. For measuring the temperature in greater heights, a Rayleigh integration lidar is ideally used. Both techniques can be combined in one system. It need not transmit at more than one wavelength. The minimum height of the measurements depends on the overlap between the transmitter and receiver of the system, whereas the measurement resolution and the maximum height of the measurements depend on the transmitted laser power and the size of the receiving telescope. With favorable transmission of the atmosphere such a combined system allows continuous temperature measurements simultaneously in the troposphere, stratosphere and mesosphere.

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# 11

## Resonance Scattering Lidar

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### 11.1 Introduction

Resonance fluorescence is the process in which the energy of an incoming photon coincides with the energy of a transition in the level scheme of an atom, ion, or molecule, and is reemitted at the same or at some longer wavelength. In lidar, we consider only those cases in which both wavelengths are the same. Resonance fluorescence is widely used for analytical purposes because of the high, narrow peaks of the resonance lines. These result in high sensitivity and high selectivity of the analytical procedure. The resonance process implies both absorption, or a loss, and reemission, or scattering of the primary radiation. Resonance *absorption* is used in differential-absorption lidar, e.g., for the measurement of mercury [1] (see Chapter 7). Resonance *scattering* has had very few applications in lidar at low and middle altitudes, for a number of reasons. First, the resonance fluorescence process is most intense on atoms and ions which have few, but very intense, lines and not so well on molecules with vibrational and rotational degrees of freedom in which the oscillator strength is distributed over broad bands with a very high number of individual, but much less intense lines. Second, fluorescence lifetimes are relatively long, limiting the temporal resolution and thus range resolution of the lidar at low altitudes where high resolution is often required. Third, nonradiative (collision) deexcitation or quenching, with the resulting loss in intensity, is important at atmospheric pressure. Finally, free metal atoms or ions for which resonance fluorescence is most intense are not abundant in the lower layers of the atmosphere.

Conditions are different, however, in the high atmosphere. In 1969 Bowman et al. [2] made the first resonance-scattering lidar observation of an atomic layer of metallic sodium (Na) in the mesopause region at altitudes between 80 and 110 km using resonance-scattering lidar. In fact, the existence of a layer of sodium atoms in the mesopause region had already been deduced from observations at twilight by Chamberlain et al. in 1958 [3]. These findings were later confirmed by a number of researchers such as Sandford and Gibson in 1970 [4], Hake et al. in 1972 [5], Mégie and Blamont in 1977 [6], later by Beatty et al. [7] and Tilgner and von Zahn [8] in 1988, and by others. In the Southern hemisphere Clemesha et al. [9] made long-term observations of the Na layer for over 15 years. In the United States, high-resolution measurements by the group led by Gardner at the University of Illinois allowed the observation of wave-like structures that were interpreted as gravity waves [10]. They also observed the horizontal structure of the Na layer in equatorial and polar areas with an airborne Na lidar [11, 12]. At about the same time, an American and a French group became successful in observing metallic species other than Na such as potassium, lithium, calcium in atomic and ionic form, and iron (K, Li, Ca,  $\text{Ca}^+$ , Fe) [13–17]. Later, these and other groups carried out simultaneous determinations of the different atoms and ions and compared the results with data from high-atmosphere observations made with different instruments [18, 19]. Later, more atmospheric quantities than just atomic densities were extracted from the data. Lidar systems for the determination of temperature from the Doppler broadening of the Na  $\text{D}_2$  lines were presented by Gibson et al. in 1979 [20], Fricke and von Zahn in 1985 [21], von Zahn and Neuber in 1987 [22], and She et al. in 1990 [23]. Later, the Na  $\text{D}_2$  Doppler shift was also used to develop a highly accurate method for determining radial wind [24]. This considerably widened the field of applications of resonance scattering lidar in the high atmosphere.

## 11.2 The Mesospheric Na Layer: Methodology and Observations

For resonance scattering lidar, wavelength tunability is clearly required for the transmitting laser. In the early days, flashlamp-pumped dye lasers were used because they provided high pulse power and were tunable over a wide range of wavelengths. They could cover, in fact, the whole visible spectrum. Dyes from the rhodamine family showed large output power

and good stability characteristics, especially at wavelengths between 550 and 600 nm, which was ideal for the Na atom with its strong D<sub>2</sub> resonance line at 589 nm. Dye lasers directly pumped by flashlamps were used in the initial stage, but this type of laser was poorly suited for long, continuous observations because of the short lifetimes of the flashlamps and of the dyes, these latter being deteriorated by the intense light of the flashlamps of which only a small spectral band was converted to useful radiation. Thus, the light from the flashlamps was later replaced by the second harmonic of the output of a Nd:YAG laser. With this monochromatic radiation of a wavelength of 532 nm, higher excitation efficiency and longer dye life were achieved which allowed better observations. Figure 11.1 shows schematically the layout of a typical lidar for Na layer observations.

What makes a resonance scattering lidar differ significantly from other lidar systems is not only that it must allow precise tuning to the resonance line of the target atom or ion, it must also maintain a narrow linewidth for the whole duration of the observation. Table 11.1 lists the technical data of the Na layer observation lidar used at Tokyo Metropolitan University. The desired laser linewidth of approximately 2.5 pm is slightly less than the width of the Na D<sub>2</sub> line. If the laser linewidth becomes wider, the resonance-scattering efficiency quickly decreases, with a corresponding reduction of the lidar signal-to-noise ratio.

At an altitude around 90 km, scattering from aerosols and even atmospheric molecules can be ignored with respect to resonance

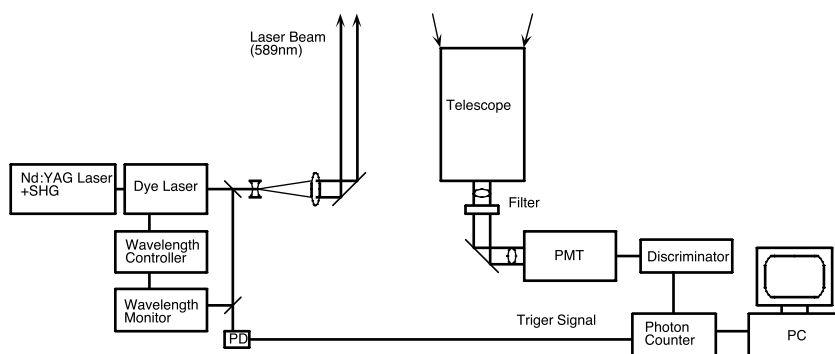


**Fig. 11.1.** Schematics of a typical resonance scattering lidar for mesospheric sodium measurements. PC data acquisition computer, PD photodiode, PMT photomultiplier tube.

**Table 11.1.** Technical data of the MTU resonance-scattering sodium lidar

|                   |                       |                           |
|-------------------|-----------------------|---------------------------|
| Transmitter:      | Laser                 | Nd:YAG SHG pumped dye     |
|                   | Wavelength            | 589 nm                    |
|                   | Pulse energy          | 100 mJ/pulse              |
|                   | Pulse repetition rate | 10 Hz                     |
|                   | Linewidth             | 2.5 pm FWHM               |
|                   | Pulse length          | 6 ns                      |
|                   | Beam divergence       | 0.2 mrad                  |
| Receiver:         | Telescope             | 60 cm diameter Cassegrain |
|                   | Field of view         | 0.7 mrad                  |
|                   | Optical bandwidth     | 3.5 nm FWHM               |
| Data Acquisition: | Type                  | Photon counting           |
|                   | Range resolution      | 100 m                     |

scattering. The lidar signal power from resonance backscattering at height  $z$  in this region can be described as

$$P(z) = \frac{P_0 A \eta T(z)^2 \rho_{\text{Na}}(z) \sigma_{\text{eff}}}{z^2}, \quad (11.1)$$

where  $P(z)$  is the lidar receiver intensity,  $P_0 = Ec/2$  the transmitting laser output if  $E$  is the pulse energy and  $c$  is the speed of light,  $A$  is the area of the receiving mirror,  $\eta$  the efficiency of the receiver system,  $T(z)$  the transmission factor from the ground to height  $z$ , and  $\rho_{\text{Na}}(z)$  the number density of Na atoms at height  $z$ . This is the familiar lidar equation. For resonance-scattering lidar, however, the cross section must be replaced with the quantity

$$\sigma_{\text{eff}} = \int g(\nu) \sigma(\nu, z) d\nu = \int g'(\lambda) \sigma'(\lambda, z) d\lambda, \quad (11.2)$$

which we call effective cross section.  $g(\nu)$  or  $g(\lambda)$  is the normalized spectral distribution of the laser power over the fluorescence line of the atom, and  $\sigma(\nu, z)$  or  $\sigma(\lambda, z)$  is the cross section distribution in the line which in principle shows a weak dependence on height  $z$ . Whether the wavenumber ( $\nu$ ) or wavelength notation ( $\lambda$ ) is used is a matter of taste. The primes indicate that the functions are not the same, but the integral is the same. Figure 11.2 shows the relationship between the laser linewidth and the effective cross section.

Figure 11.3 is a plot of the raw lidar data of a height profile with echo from a layer of Na atoms with maximum centered around 90 km height. Signal contributions from aerosols can be totally ignored. For

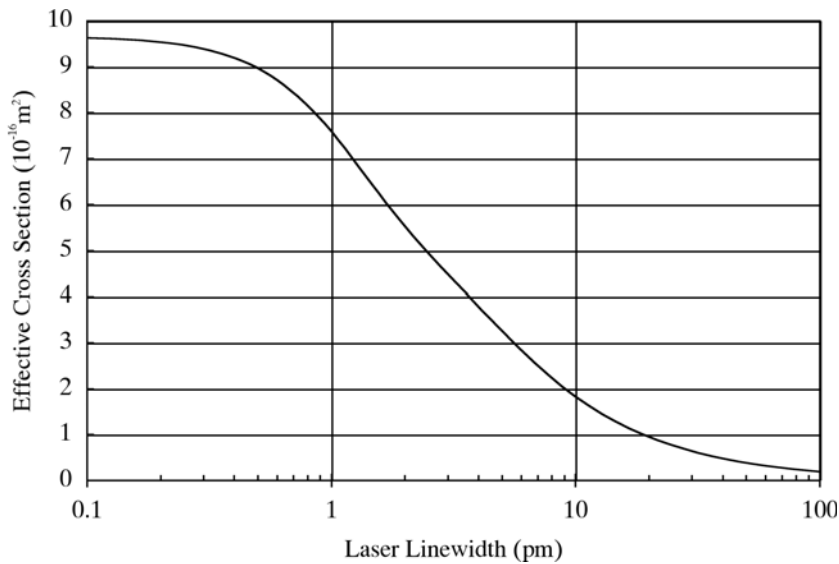


**Fig. 11.2.** Effective resonance cross section versus laser linewidth.

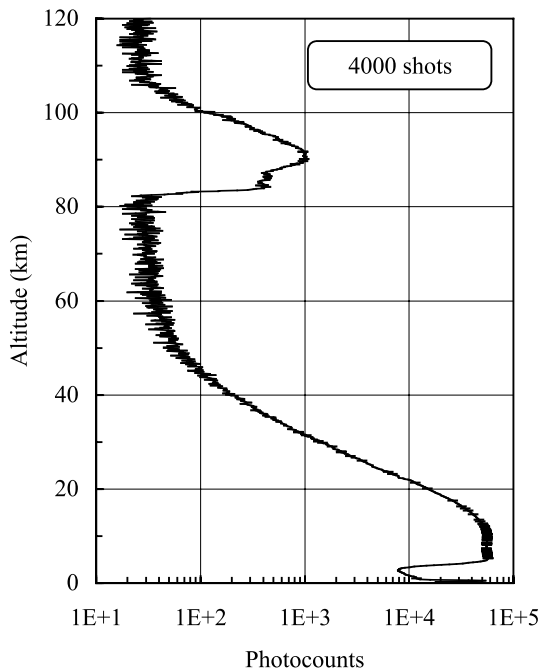


**Fig. 11.3.** Raw data of a sodium lidar profile (4000 shots).

the calculation of Na atomic density from the raw data, the signal at an altitude  $z_R$  is used at which molecular scattering is fully present and constitutes the only contribution to the lidar signal. This is normally an altitude around 30 to 35 km. This signal is given by

$$P(z_R) = \frac{P_0 A \eta T^2 \rho_M(z_R) \sigma_M}{z_R^2}. \quad (11.3)$$

$\rho_M(z_R)$  is the number density of atmospheric molecules at altitude  $z_R$  and  $\sigma_M$  is their Rayleigh scattering cross section. With the known density  $\rho_M(z_R)$  the number density of the Na atoms is then easily determined by taking the ratio of Eqs. (11.1) and (11.3). The result is

$$\rho_{Na}(z) = \frac{P(z) \rho_M(z_R) \sigma_M z^2}{P(z_R) \sigma_{eff} z_R^2}. \quad (11.4)$$

In doing so, we had to use the assumption that the additional absorption by the sodium atoms in the transmission term can be neglected. This assumption is justified.

Figure 11.4 shows as an example the distribution of Na atom densities taken at Tokyo Metropolitan University on the night of 14 to 15 January 2004. It displays one Na atomic density profile every 8 minutes, from sunset to sunrise.

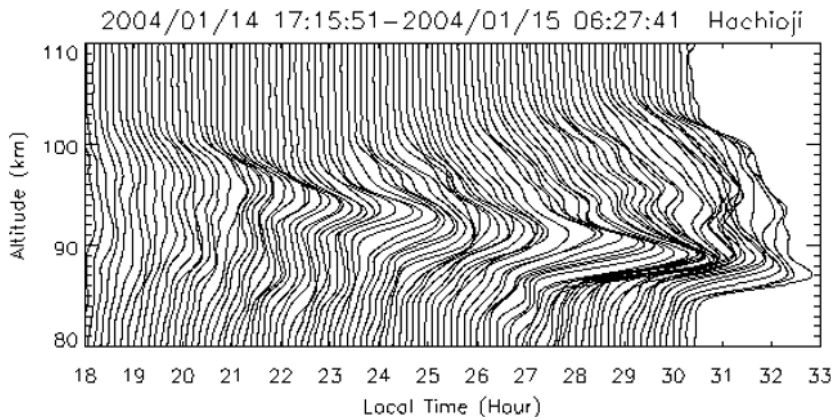


**Fig. 11.4.** Time series of mesospheric sodium layers observed at Tokyo in the night of 14 to 15 January 2004. Concentrations are in arbitrary units.

Daytime observations also are of considerable interest. Because of the intense skylight background and resulting low signal-to-noise ratio, such daytime measurements are not easy to make. Gibson and Sandford [25] used a Fabry–Perot etalon as a narrowband filter and were the first to present Na density profiles during daytime. In 1982 Clemesha et al. [26] and Granier and Mégie [27] and in 1987 Kwon et al. [28] continued such observations. However, there are not many cases of continuous observations reported because of the difficulties with etalon control and stabilization. In 1996, Chen et al. [29] successfully carried out daytime observations of the Na atomic layer in the mesopause region with relatively simple equipment, viz., a Faraday band-pass filter.

The filter consists of a Na atomic vapor cell in a magnetic field between crossed polarizers. The magnetic field Zeeman splits the energy levels, resulting in separate absorption lines for left and right circularly polarized light. We can consider the filter transmission in terms of circularly polarized light outside, between, and at the absorption lines. Outside these lines, the filter can be regarded as a Faraday rotator that uses Na vapor as the magneto-optic material inserted between crossed polarizers. The Na vapor exhibits rotary power only in the immediate vicinity of an absorption line, providing the 90-degree turn needed to pass the second polarizer. A simple peaked transmission spectrum is obtained when the filter parameters such as cell temperature and magnetic field are adjusted to provide a maximum rotation of 90 degrees. Away from the absorption line, the filter provides an out-of-band rejection determined by the extinction ratio of the crossed polarizers. In this way it becomes possible to take high-quality lidar data continuously day and night.

Among the geophysical questions directly related with Na atom abundance, the origin, distribution and wave-like behavior have dominated the discussions until now. As to the origin, diurnal and seasonal variations as well as the latitudinal distribution [30] give much insight into the effects responsible for the Na atom concentrations, but there is as yet no closed model of Na atom generation and transport, and the discussion of the related processes is far from being closed.

Gravity waves for which Na layers from the beginning of these investigations have been used as a near-ideal tracer, reveal the complex dynamics of the upper atmosphere, and both measurements and models continue to be an important subject in high-atmosphere research [31].

One of the most interesting and least understood phenomena in the mesopause region is perhaps the occurrence of sporadic Na layers (Na<sub>s</sub> layers). Na<sub>s</sub> layers are characterized by large density enhancements

in narrow altitude ranges, less than a few kilometers thick.  $\text{Na}_s$  layers were first observed at low and high, but not at mid-latitudes [32–36]. However, in 1995 observations by Nagasawa and Abo [37] indicated that the occurrence rate of  $\text{Na}_s$  layers at Hachioji, Tokyo ( $35.6^\circ\text{N}$ ,  $139.4^\circ\text{E}$ ) (Fig. 11.5) can be comparable with those at low and high latitudes [32, 33, 38]. The formation mechanism of  $\text{Na}_s$  layers is actually under intense discussion. The high correlation observed between the occurrence of  $\text{Na}_s$  and sporadic E layers ( $E_s$ ), which are the thin layers of enhanced ionization in the ionospheric E region, led to the hypothesis that the neutralization of a  $\text{Na}^+$  (sodium ion) reservoir in the  $E_s$  layers was a source of large quantities of neutral Na atoms [36, 39, 40]. However, a recent observation at Arecibo Observatory showed that the appearance of an electron layer followed that of the correlated  $\text{Na}_s$  layer at its peak-abundance altitude [41]. Von Zahn et al. [42] also suggested that the  $\text{Na}_s$  layers at high latitude could originate from the release of Na by dust and smoke particles under the effect of energetic-particle bombardment. Even if this is correct, it remains difficult to explain the formation of  $\text{Na}_s$  layers at low latitudes by this mechanism [38]. Dynamical effects

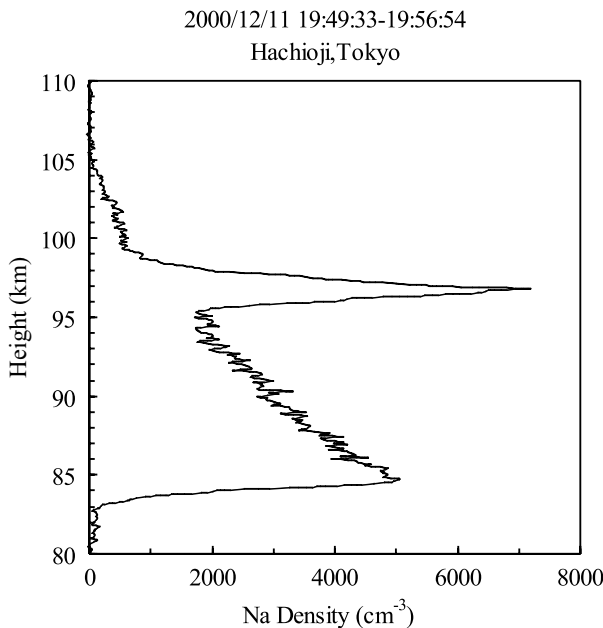


**Fig. 11.5.** Sporadic sodium layer observed at Tokyo on 11 December 2000.



associated with tides, gravity waves, and turbulence may contribute to the formation of Na<sub>s</sub> layers [43,44]. The significant difference in the occurrence rates at high and low from those at mid-latitudes has raised new questions as to the formation mechanisms of Na<sub>s</sub> layers. Nagasawa and Abo suggested in 1995 that the occurrence of Na<sub>s</sub> layers depends on geomagnetic rather than geographic latitude [37].

### 11.3 Observations of Other Metallic Layers

In addition to sodium, various other metal atoms and ions have also been considered as targets of mesosphere lidar observations. The most important ones are listed in Table 11.2. The Haute-Provence observation station in France (44°N) succeeded in observing metallic species such as K, Li, Ca, Ca ion, and Fe one after another, after Felix et al. in 1973 [13] and Mégie et al. in 1978 [14] had first seen and measured atomic potassium in the mesopause region using a ruby-laser-pumped dye laser. Suitable resonance lines are the ones at 769.9 and 766.5 nm. In practice it turns out that the weaker line (at 769.9 nm) is the better choice because the 766.5-nm line overlaps with an absorption line of the oxygen molecule. By tuning a flashlamp-pumped dye laser with 800 mJ output pulse energy to the Li resonance line of 670.8 nm, Jegou et al. [15] succeeded in observing Li at one thousandth the density of Na and in measuring the <sup>6</sup>Li/<sup>7</sup>Li isotope ratio. They measured the isotopic ratio in meteor showers in order to obtain the cosmological isotopic ratio and compared it with the isotope ratio of terrestrial lithium. Granier et al. [16] could measure the atomic density of calcium with a dye laser at a wavelength of 422.7 nm

**Table 11.2.** Resonance lines of mesospheric metallic species used in resonance scattering lidar [45]

| Metallic species | Resonance wavelength in air (nm) | Backscatter cross section (m <sup>2</sup> sr <sup>-1</sup> ) |
|------------------|----------------------------------|--------------------------------------------------------------|
| Fe               | 371.993                          | $8.15 \times 10^{-18}$                                       |
| Ca <sup>+</sup>  | 393.366                          | $1.12 \times 10^{-16}$                                       |
| Ca               | 422.673                          | $4.17 \times 10^{-16}$                                       |
| Li               | 670.776                          | $1.12 \times 10^{-16}$                                       |
| Na               | 588.995                          | $7.78 \times 10^{-17}$                                       |
| K                | 769.897                          | $7.51 \times 10^{-17}$                                       |

pumped by the third harmonic of a Nd:YAG laser. The same authors measured Ca ions with radiation of 393.4 nm wavelength [17]; to this end they mixed dye laser radiation of 624 nm with the 1064-nm Nd:YAG-laser fundamental. They finally succeeded in the measurement of Fe atomic density by mixing dye laser radiation of 572 nm and the Nd:YAG-laser fundamental of 1064 nm to produce 372.0 nm. Bills and Gardner [46] similarly measured Fe density in Illinois (40°N) using excimer-laser pumped dye lasers capable of high average output; their results were 5 to 8 times the Fe atomic densities observed by Granier et al. [17].

Gardner et al. [18] simultaneously measured Na, Fe, and  $\text{Ca}^+$  densities and temperature (described in the following section), and Qian and Gardner [47] did a simultaneous observation of Ca and Na densities and temperature. Examples of their measurements are shown in Fig. 11.6. Alpers et al. [19] have been successful in simultaneously measuring the density of Ca and  $\text{Ca}^+$ . There are more examples of recent simultaneous observation with metallic atoms. Few of these use dye lasers any more. Instead, tunable solid-state lasers such as alexandrite and  $\text{Ti:Al}_2\text{O}_3$  (or TiSa, for titanium-sapphire) lasers which are easier to use, particularly on mobile platforms, have taken over; e.g., Eska et al. [48] demonstrated the feasibility of measurements of latitudinal variations of K density with an alexandrite-laser-based lidar installed on board a vessel.

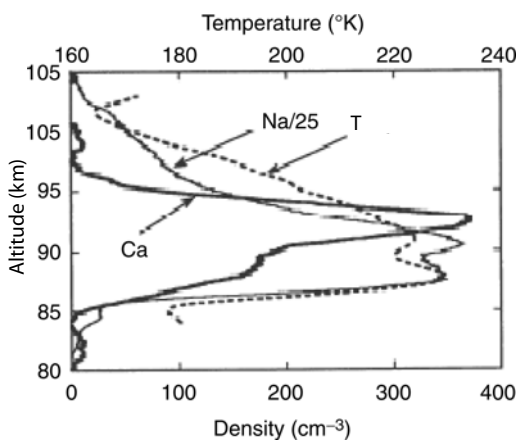


**Fig. 11.6.** Simultaneous common volume measurements of Ca density, Na density, and temperature (T) profiles. From Qian and Gardner, 1995 [47].

## 11.4 Measurements of Temperature and Wind with Resonance-Scattering Lidar

As shown in Fig. 11.7, the Na D<sub>2</sub> line undergoes Doppler broadening with atmospheric temperature; by measuring this broadening the temperature of the sodium layer can be determined. As the average number of collisions with atmospheric molecules at 90 km altitude is  $10^4/\text{s}$ , it can be assumed that the Na is in local thermal equilibrium with its surroundings or, in other words, that its temperature is the same as that of the sodium.

The first to succeed in measuring the temperature of the sodium layer using resonance scattering lidar were Gibson et al. in 1979 [20]. Using a narrowband laser (laser linewidth approximately 100 MHz) they took resonance-scattering data at eight wavelengths within the Na D<sub>2</sub> line and fitted the measured data to the theoretical values of the Doppler-broadened line. However, their measurement range and height resolution were limited so they could only determine the average temperature in the central area of the sodium layer. Neuber et al. [49] later continuously measured the temperature in Andoya (69.3°N) using a similar method with excimer-laser-pumped dye lasers. Between the altitudes of

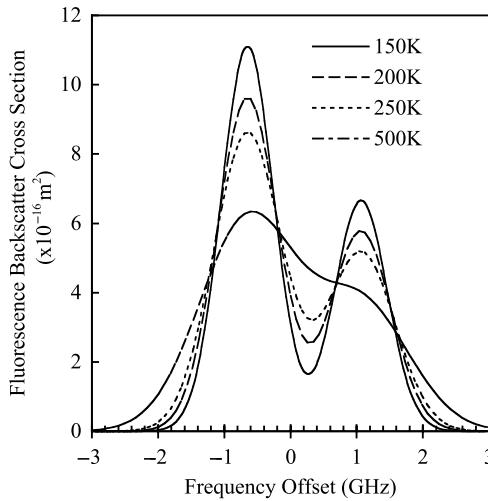


**Fig. 11.7.** Na D<sub>2</sub> Doppler-broadened fluorescence spectrum plotted as a function of frequency for four temperatures.

85 and 95 km and at a height resolution of 1 km, the accuracy of their temperature measurement was  $\pm 5$  K.

She et al. [23] developed a method to measure temperature with higher accuracy. The principle of measurement has been described in detail by Bills et al. [50]. In order to obtain spectrally narrow laser emission, the authors injected radiation from a narrowband ring dye laser into a Nd:YAG-pumped pulsed dye laser. The frequency jitter obtained in this way was 1 MHz or less. Figure 11.8 is a diagram of their laser system. Their measurement device is also characterized by the utilization of Doppler-free saturated-absorption spectroscopy in a Na cell to alternately tune the lidar's optical transmission with high precision to the two most appropriate wavelengths within the Na D<sub>2</sub> line spectrum. Figure 11.9 shows the Na fluorescence spectrum from the Na vapor cell that is used. The spiked area in the spectrum is used as the tuning point. The final shape of the laser spectrum is then monitored with a Fabry–Perot etalon and corrected to maintain measurement accuracy. As a result, 5-minute measurements between 80 and 105 km and with 1 km range resolution yielded a temperature accuracy of  $\pm 1$  K near the sodium peak of the Na layer and  $\pm 3$  K elsewhere.

Using this technology in connection with the Faraday band-pass filter described in Section 2, daytime and nighttime measurements of the temperature were obtained. The results were quite different from those of conventional models. She et al. [24] further improved the Na temperature measuring device to measure the radial velocity with which the sodium layer moves, or wind. She et al. [51] provided eight-year climatology data of temperature profiles in the mesopause region. Temperature

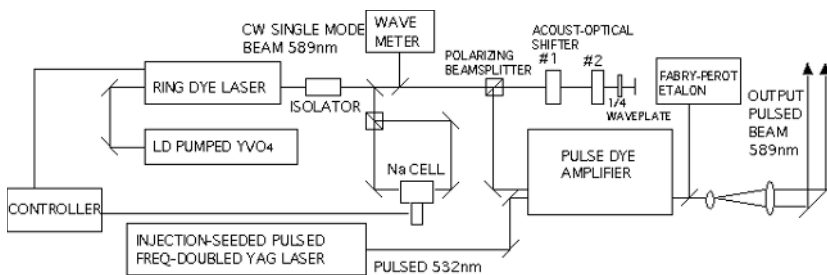


**Fig. 11.8.** Block diagram of the Na temperature lidar transmitter.

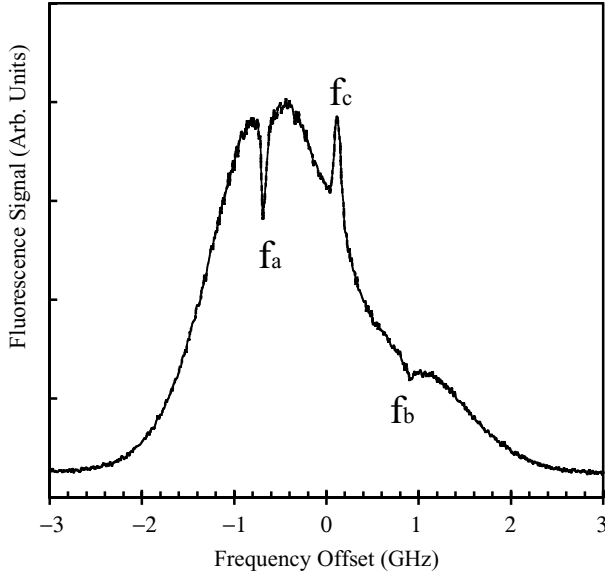


**Fig. 11.9.** The measured Na vapor cell fluorescence spectrum. The labels denote the Doppler-free feature at the  $D_2a$  peak ( $f_a$ ), crossover resonance ( $f_c$ ), and  $D_2b$  peak ( $f_b$ ).

contours support a two-level thermal structure. Examples of temperature measurements are shown in Fig. 11.10 [52].

Von Zahn and Höffner [53] used another element, potassium, to measure temperature. They employed a different measurement scheme with tunable solid-state lasers that are easier to maintain and operate than dye lasers.

Kawahara et al. [54] succeeded in temperature measurements over the Syowa station ( $69^\circ\text{S}$ ,  $39^\circ\text{E}$ ) in Antarctica using a Na resonance line generated by sum-frequency mixing of two injection-seeded pulsed Nd:YAG lasers.

Gelbwachs [55] proposed to measure mesopause temperatures by determination of the Boltzmann factor instead of the Doppler broadening of a resonance line, preferably on iron. Gardner et al. [56] applied the method in an airborne lidar to the measurement of Leonid meteor trails and for observations over Antarctica [57]. The advantage of this method is that laser wavelength tuning need not be extremely accurate; however, there is the disadvantage that the resonance scattering cross sections of the two wavelengths used in the measurement (372 nm and 374 nm) are small and the echoes are weak.

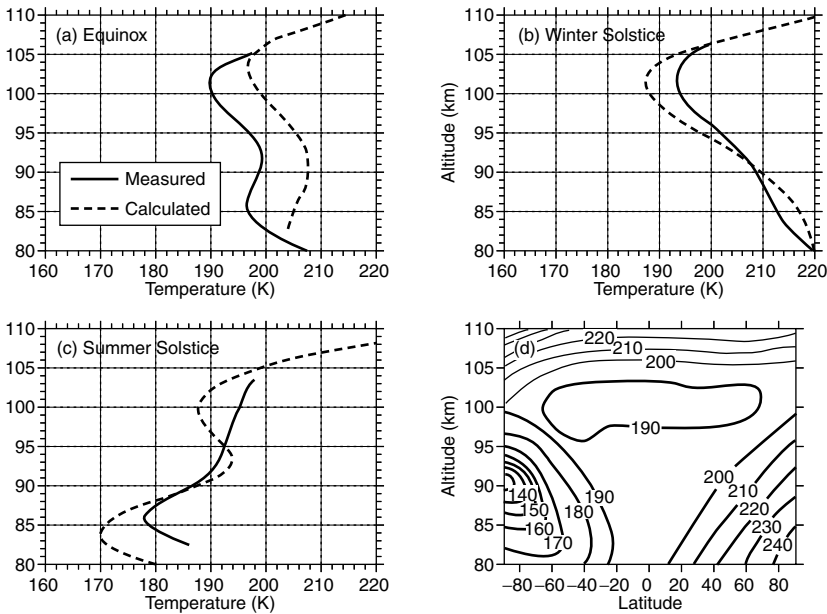


**Fig. 11.10.** Calculated and measured nighttime averaged temperature profiles over Fort Collins, CO ( $41^{\circ}\text{N}$ ,  $105^{\circ}\text{W}$ ) for (a) equinox, (b) winter, (c) summer, along with calculated altitude-latitude plots of zonal mean temperature contours in the mesopause region for December solstice solar cycle minimum geomagnetic quiet conditions, in (d). From She et al. [52].

## 11.5 Summary and Future Prospects

In addition to the possibility of measuring the primary target of resonance-scattering lidar systems, i.e., the layers of metal atoms and ions in the mesopause region, now other parameters such as temperature and wind and their temporal and spatial distribution can also be determined. This greatly increased the range of investigations in the high atmosphere that can be tackled: dynamics, climatologies, meteoric material and its nature, quantity, frequency and time distribution of occurrence, and origin, to name just a few.

Of all competing technologies for measurements in this part of the atmosphere, resonance-fluorescence lidar combines such properties as good accuracy, specificity, resolution, independence from many perturbing conditions that affect other measurement schemes, safe operation from the ground, and low cost. It is thus ideally suited to supplement investigations made with other equipment that do not share these assets.

Examples are the middle and upper atmosphere (MU) radar [58], medium frequency (MF) radar [59], and CCD cameras [60]. This is certainly one reason why resonance-scattering lidars are being built and used in various parts of the world.

Resonance-scattering lidars, despite all their desirable features, suffer from two disadvantages. One is the fact that, because of ozone absorption, they cannot use excitation wavelengths much shorter than 300 nm; even high-power systems with large receiver telescopes will not extend that limit dramatically. The other disadvantage is that they do not work in cloudy weather. Both shortcomings are overcome when the systems are taken on high-flying planes or, better, satellites [61]. Work is in preparation or under way for the development of highly reliable, full solid-state Na lidar systems that meet the requirements for unattended operation [62, 63].

These technological improvements, however, are not the only direction in which resonance-fluorescence lidars develop. There are new physical challenges as well. An example is the work by Brinksma et al. [64] who were the first to succeed in the observation of OH molecules in the mesosphere using resonance-scattering lidars with excimer lasers. It might thus be possible to extend the range of target substances to other fluorescent molecules. Finally, as an example of application in areas outside the field of atmospheric science, laser guide stars are being developed in which a system similar to a resonance-scattering lidar targeting mesospheric Na layers is used. The resonance scattering from the Na layer can provide the beacon for adaptive-optics compensation of atmospheric distortion [65].

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## Doppler Wind Lidar

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### 12.1 Introduction

The change of perceived frequency of radiation when the source or the receiver move relative to one another is a well-known phenomenon. First described by Austrian physicist Christian Doppler (1803–1853) for acoustic waves, it occurs for electromagnetic waves as well. If the change of frequency can be measured, the relative speed of the source with respect to the receiver can be determined, provided the group velocity of the radiation in the respective medium is known. As the speed of light in air and vacuum has been known with high accuracy, the optical Doppler effect lends itself ideally to the remote measurement of the speed of very distant or otherwise uncooperative objects. If the object does not move directly toward or directly away from the observer, then the use of the optical Doppler effect clearly yields the component of the speed of the object along the line of sight.

It is obvious that for a velocity measurement the object must emit electromagnetic radiation. This is the case for stars and galaxies; perhaps the most spectacular application of the optical Doppler effect was the determination of the shift of light from distant stars, all toward longer wavelengths, leading to our present notion of an expanding universe. Because the relative shift of optical frequencies,  $\Delta f/f$ , is proportional to  $v/c$ , the ratio of the velocity  $v$  of the object to the speed of light  $c$ , and because very distant stars move away fast, these measurements were comparatively easy to make.

Velocity determinations on Earth and in the Earth's atmosphere are more difficult for two reasons. First, the objects whose speed is to be

measured must be made to emit radiation. This can be done, e.g., by illumination. Second, the shift of the return radiation with respect to the transmitted radiation must be determined. Velocities of interest on Earth vary greatly with object and purpose. The movement of air masses, e.g., is interesting at velocities of about 0.1 to 100 m/s which, relative to the speed of light of  $3 \times 10^8$  m/s, amounts to a fraction of roughly 3 parts in  $10^{10}$  to 3 parts in  $10^7$ . This is not easy to measure unless very narrow spectral lines and highly sophisticated equipment are used.

Although optical Doppler measurements have a multitude of terrestrial applications such as the determination of the speed and vibrations of moving parts in traffic, in industrial production, in machine shops, etc., this chapter is exclusively devoted to the measurement of the movement of atmospheric air masses, or wind and turbulence, from the observation of aerosols. Compared with other Doppler measurements, Doppler wind measurements have the additional problems that the illumination of the air even with powerful sources yields very weak return signals and that the return signals must be analyzed not just for wavelength, but for distance as well. Following this Introduction, Section 12.2 will briefly recall the notations and formulas that will be used in connection with optical Doppler wind lidar. In Section 12.3 different schemes for remote measurements of the wind vector are presented. In Section 12.4 the wavelengths to be used, the different detection schemes and the various scan techniques for Doppler lidar are discussed. Section 12.5 shows several applications, with main emphasis on heterodyne wind lidar, and Section 12.6 concludes this chapter with a number of new areas in which optical Doppler wind lidar may gain importance in the future.

## 12.2 The Optical Doppler Effect

Light, unlike sound, is not “advected” by some medium. In the optical Doppler effect, there is therefore no distinction between the case of the moving transmitter and the moving receiver, or both transmitter and receiver moving in a medium. If the emitted light has wavelength  $\lambda_0$  and frequency  $f_0 = c/\lambda_0$  and the relative speed along the line of sight is  $v$ , then the observed frequency is

$$f = f_0(1 + v/c). \quad (12.1)$$

Air and aerosols, however, normally do not emit light, but for a measurement of their speed are illuminated by light from the lidar transmitter.

If that light has frequency  $f_0$ , then its apparent frequency on the aerosol particle is given by Eq. (12.1). Clearly, the light is reemitted, or back-scattered, at this frequency, which then, because the particle is moving while scattering, is detected by the lidar receiver as being shifted to frequency

$$f = f_0 + \Delta f = f_0(1 + 2v/c). \quad (12.2)$$

We define the particle (or wind) velocity in such a way that a movement *toward* the lidar which leads to a *positive* frequency shift is characterized by a *positive* line-of-sight velocity, and vice versa. Instead of the line-of-sight velocity  $v_{\text{LOS}}$  or velocity component along the line of sight, we occasionally use the term “radial velocity”  $v_r$  or radial component of a velocity vector that is not parallel to the line of sight.  $v_{\text{LOS}}$  and  $v_r$  are fully synonymous, with the same sign convention.

Now the collective movement of air masses which we call wind is superimposed by the individual, thermal, random movement of the molecules. These normally move much faster than the wind speed and so much the faster the higher the temperature. The relative shift of their velocity distribution with the wind speed is therefore small. Aerosol particles, because of their higher mass, move more slowly at the same temperature and have therefore a narrower velocity distribution. They are shifted by the same amount, but relative to its width this shift is much larger and amenable to measurement. The situation is schematically shown in Fig. 12.1.

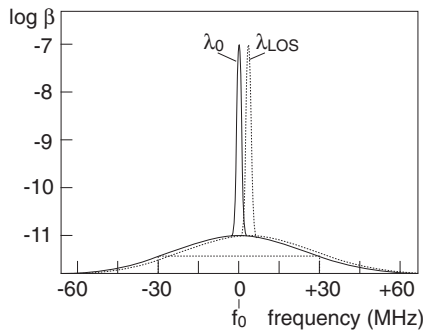


**Fig. 12.1.** Schematic representation of the original (solid) and wind-shifted (dotted) frequency distributions. If there are aerosols present, a narrow spike is superimposed onto the broad molecular peak. The return-signal frequency is shifted here toward higher values, indicating that the wind comes toward the lidar. At  $10.59\text{ }\mu\text{m}$  wavelength, the 3-MHz shift corresponds to roughly 20 m/s.

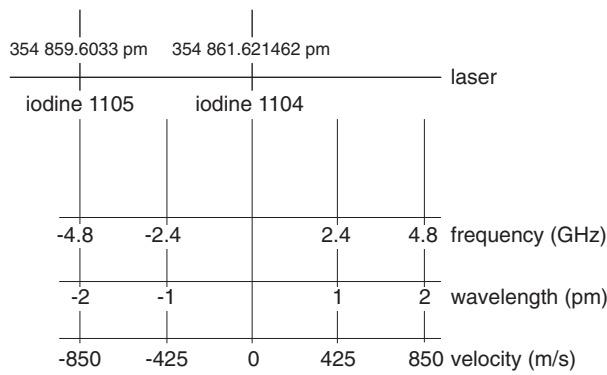


**Fig. 12.2.** Relation of wavelength, frequency and wavelength shift with wind speed for Doppler wind lidar measurements with a frequency-doubled Nd:YAG laser stabilized to the iodine 1104 atomic transition. Note that wind speed scale is a factor  $>1000$  too coarse for most practical applications.

To give an idea how small wind-induced shifts are relative to the absolute wavelengths or frequencies and to demonstrate that lidar transmitters and receivers must be controlled to sub-picometer accuracy, Fig. 12.2 shows how wind velocities (bottom scale) translate into wavelength shifts and frequency shifts (center scales) if light from a frequency-tripled Nd:YAG laser is used. The top scale shows the positions of two iodine lines (often designated as # 1105 and # 1104) to which the laser can be stabilized [1]. In direct-detection lidars and in the field of optical space communication it is necessary to stabilize the laser with respect to the filters in front of the detectors.

### 12.3 Brief Overview of Wind Lidar Measurement Schemes

Pulsed Doppler wind lidar is not the only method for the remote optical measurement of wind speed. There are other optical methods that shall be briefly presented here for the sake of completeness. They will not be treated in detail because their application is limited to short range due the very principle of the measurement, because the method is relatively new and its technical implementation is complicated and still in its infancy, or because they are described elsewhere in this book.

### 12.3.1 Crosswind Determination by Pattern Correlation

Cloud droplets and aerosol particles are not distributed homogeneously in the atmosphere. Smokestack plumes and clouds show patterns easily recognized with the naked eye. If images of such patterns are taken at two points in time,  $t_1$  and  $t_2$ , and if the geometric parameters such as distance, angle of observation, and imaging scale are known so that the two-dimensional pattern  $H(x, y)$  of the object can be determined from the images, then it is sufficient to find those two values  $(\xi, \eta)$  by which the second image must be shifted to give maximum similarity with the first. Or in mathematical terms, we need to determine those values  $(\xi, \eta)$  that maximize the cross-correlation coefficient of the two images:

$$Q(\xi, \eta) = \iint H(x, y, t_1)H(x - \xi, y - \eta, t_2) dx dy = \text{maximum.} \quad (12.3)$$

The (two-component) velocity vector in the plane perpendicular to the line of sight (normally the horizontal plane) is then given by the simple relation

$$\mathbf{u}_{\text{hor}} = \frac{1}{t_2 - t_1}(\xi, \eta). \quad (12.4)$$

We shall not dwell on the fact that instead of the convolution of Eq. (12.3) fast Fourier transform algorithms are the preferred method to obtain the shift parameters  $(\xi, \eta)$ .

The method can be used in the daytime in an entirely passive way, but only for the altitude at which the receiver sees a smokestack plume or cloud patterns. By selecting an appropriate wavelength it is possible to make the method particularly sensitive for one type of object: uv for  $\text{SO}_2$ , visible for clouds and black smoke, ir for very warm plumes. Outside plumes and for nighttime measurements the objects can be illuminated. More efficient than the use of searchlights have been scans of the scene with pulsed lasers. By using a time-resolving detector, height-resolved, and, that is, genuine lidar measurements are possible with this technique. Using a pulsed ruby laser, in fact, Sroga et al. [2] could measure vertical profiles of the horizontal wind vector between 120 and 600 m height as early as 1980. In those days scanning was not possible fast enough for the scan of a set of pictures to be completed in a time  $\ll t_2 - t_1$ , but Sasano et al. [3] soon developed a correction algorithm for the resulting error. Today, with much more powerful equipment and much faster

computers available, the technique has been developed to a high degree of sophistication (cf. Chapter 5 of this book).

### **12.3.2 Laser Time-of-Flight Velocimetry (LTV)**

If two laser beams are focused at some distance from the ground and close to each other and an aerosol particle crosses both foci, then two flashes can be seen, the interval between the flashes being indicative of the speed of the particle. Because aerosol particles are advected with the wind, the wind speed can be obtained in this way.

It is not obvious that, for a given wind direction, particles will cross both foci unless their connecting line happens to coincide with the wind direction. If the distance between foci is sufficiently small, however (on the order of 1 mm), then there is sufficient scatter in the direction of the particle trajectories to provide useful results. The depth of the focal volume is in any case much larger than its lateral dimensions, so a vertical component of the particle velocity vector is not critical. By rotating the apparatus  $90^\circ$  or by some other measure [4] the perpendicular component of the wind can be determined in the same way. From both results the wind direction and wind speed can then be inferred. The technique has been tested with argon ion lasers of 500 and 200 mW cw power at wavelengths of 514 and 488 nm; maximum range was 70 and 100 m, respectively [4, 5]. The theory of the method was treated in great detail by She and Kelley [6]. In spite of its simplicity, it has not nearly found the same degree of acceptance as a similar technique, laser Doppler velocimetry (see below).

### **12.3.3 Laser Doppler Velocimetry (LDV)**

For moderate distances the horizontal component of the wind vector can also be determined by a method known as laser Doppler velocimetry (LDV). Its principle is similar to that of LTV in that the speed of aerosol particles is also inferred from the time between successive flashes the particles emit when crossing areas of high and low optical intensity. The difference is that in LDV not just two foci, but a large field of interference fringes is used for illumination. Such fields can be obtained from a laser if its beam is divided in two and the fractions are transmitted by different optics oriented at a small angle with respect to one another. The resulting interference pattern acts as a periodic field of regions with high and low intensity. Particles that cross it manifest themselves by

periodic scattering of light with a frequency that is proportional to their speed.

The technique can also be used in two and three dimensions if the volume of interest is illuminated by light of different color and one color-sensitive detector is used for each dimension. For two dimensions the geometry can be relatively simple, with the axes of the two pairs of beams parallel and the connecting lines between each pair of transmitters perpendicular to one another. If the third dimension is required, the arrangement gets more complicated.

LDV is a method for the measurement of the velocity component *transverse* to the laser beam axis. The mathematical formalism, however, results in formulas that are formally identical with those developed in Section 12.2, hence the expression laser *Doppler* velocimetry. Details of the method are found, e.g., in [7] and [8].

### 12.3.4 Continuous-Wave Doppler Lidar

Continuous-wave lasers have also been used for the measurement of the longitudinal, or line-of-sight, component of the wind vector using techniques similar to those for genuine pulsed Doppler wind lidar as will be described in the remaining sections of this chapter. In cw Doppler wind lidar depth information is obtained by purely geometric means. If the detection system is focused to distance  $x$ , then roughly half of the backscatter signal is generated in a depth range

$$\Delta x = \frac{4x^2\lambda}{A} \quad (12.5)$$

if  $A$  is the detector area and  $\lambda$  the wavelength [9]. For a telescope diameter of 500 mm the depth uncertainty at a distance of 100 m is thus only 2 m which is quite good, but 200 m at 1000 m distance which is unacceptable. Although the applicability of the method has been demonstrated not just for ground-based [10], but for airborne systems as well [11–13], its use in practical applications is limited to distances well below 1000 m.

### 12.3.5 Pulsed Doppler Lidar

The restrictions of methods described in Subsections 12.3.1–12.3.4 are not given for pulsed Doppler lidar. If the term “Doppler lidar” is used in connection with wind measurements, it is implicitly understood that pulsed Doppler lidar is what the speaker has in mind. Pulsed Doppler



lidar has so much greater capabilities for the truly remote measurement of air movements that all other optical techniques of measuring air speed are practically confined to applications in the laboratory, in the machine shop, in production facilities, in wind tunnels, etc., but are hardly ever used in the free atmosphere.

## 12.4 Doppler Wind Lidar Detection and Scan Techniques

### 12.4.1 Wavelength Considerations

In Doppler wind lidar the laser wavelength can be chosen at random. However, because the aerosol contribution to the return signal is much better suited for frequency analysis than the molecular signal, the choice of the wavelength to be used will depend on the expected magnitude of the return signal and the expected ratio of aerosol-to-molecular backscatter. The molecular signal is proportional to  $\lambda^{-4}$ , the aerosol signal, depending on wavelength range and particle properties, to something between  $\lambda^{-2}$  and  $\lambda^{+1}$ . Thus, even if the aerosol return decreases with an increase in wavelength, the molecular “background” decreases much faster so the aerosol-to-molecular backscatter ratio gets more favorable.

Figure 12.3 shows simulated spectra (see also Section 6.1) of the return signals obtained with a spaceborne lidar with frequency-tripled (355-nm) Nd:YAG laser for two height bins. A high-resolution system which uses the narrow aerosol peak will work well for medium heights of 2–3 km, but not at high altitudes between 9 and 10 km where fewer aerosol particles are present. Figure 12.3 also shows that despite the shorter distance to the lidar, much fewer photons come back from the higher than from the lower interval because of the decrease of air density with height.

### 12.4.2 Detection Techniques

#### Direct Detection

As can be seen from Fig. 12.3, situations may occur in which the molecular part of the backscatter spectrum must be used. This part is frequently referred to as the Rayleigh component (as opposed to the aerosol part, which is often called Mie component) and approximated by a gaussian. The gaussian distribution is a good approximation because thermal motion of the molecules, not the effects of collisions, is the dominating

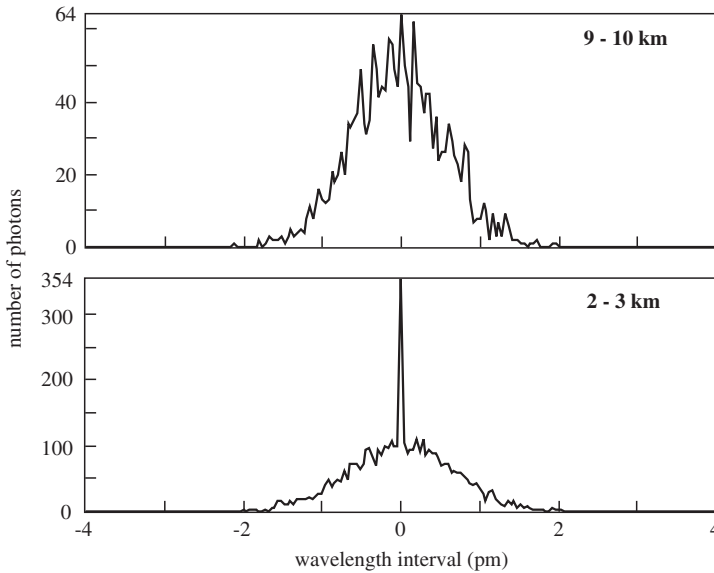


**Fig. 12.3.** Simulated return spectra for a spaceborne lidar operating at a wavelength of 355 nm. Spectra from 2 to 3 (bottom) and 9 to 10 km height (top) for zero line-of-sight wind speed.

source of line broadening and because collective effects responsible for Brillouin scattering cannot be neglected for accurate wind estimates [14].

For the determination of the center of the distribution from which the wind speed  $v$  is obtained by inversion of Eq. (12.2), different techniques are available. One is the use of a high-dispersion multichannel spectrometer that yields the whole distribution which is then submitted to a least-squares gaussian fit. Another is the use of filters such as Fabry–Perot interferometers or etalons [15–18]. Because the shifts are so small these filters must be operated on the edge of the transmission curve where the change of filter transmission with wavelength is maximum. Because the dynamic range of the signals is so large the filters must be as nearly identical as possible, except for the center wavelength of the transmission curves. For high transmitted intensity the transmission curve should cover as much as possible of the respective half of the lidar return signal. To avoid perturbations from the central Mie peak, the transmission function should be zero at line center even under conditions of nonzero wind when the Mie peak shifts from its zero-wind position.

The principle of the technique is depicted in Fig. 12.4. We call the power, or energy, or number of photons counted after transmission through filter 1 and filter 2,  $A$  and  $B$ . The difference  $A - B$ , normalized to their sum  $A + B$ , or

$$q = \frac{A - B}{A + B} = f(v), \quad (12.6)$$

depends in a unique way on wind speed  $v$ . Ideally, when this function  $f$  is inverted, it could directly yield the line-of-sight speed  $v$ . However, the measured values  $A$  and  $B$  are contaminated by background  $a$  and  $b$ . Under favorable experimental conditions this background can be measured [15] and subtracted from the apparent values  $A$  and  $B$ . The situation is more critical if the Mie peak has an intensity not negligible with respect to that of the Rayleigh peak. Let us call its contribution to the apparent values  $A$  and  $B$  for zero LOS velocity ( $v = 0$ )  $a_1$  and  $b_1$ . When the whole distribution and the Mie peak with it begin to shift to some value  $v < 0$  (which corresponds to a frequency shift  $< 0$  and a wavelength shift  $> 0$  as indicated by the dotted line in Fig. 12.4,  $a_1$  will decrease and  $b_1$  will increase, but not by the same amount. The following parameters must thus be available for use of a direct-detection Doppler lidar:

- the difference between laser wavelength and etalon transmission line center wavelengths,
- the background values in channels  $A$  and  $B$ ,

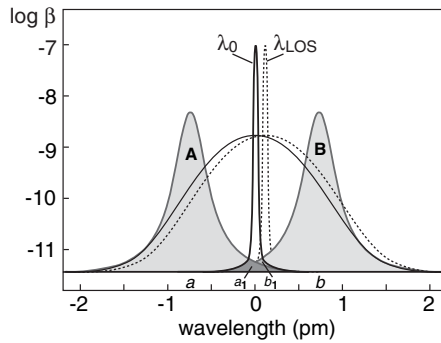


**Fig. 12.4.** Schematic representation of spectrum components in a direct-detection Doppler wind lidar.

- the temperature, and
- the aerosol scattering ratio.

The latter three parameters change dramatically with altitude, so height profiles are needed. For practical use the background must be known to an accuracy of 1–2%, the temperature which is necessary for the determination of the width of the gaussian to 1 K, and the aerosol scattering ratio, i.e., the ratio of the intensities of the aerosol backscatter to the sum of molecular and aerosol backscatter, to about 5%.

The use of direct-detection Doppler wind lidar thus represents a considerable challenge. And yet a technical implementation of a uv direct-detection lidar has been proposed by Schillinger et al. [19] which is to use the double-edge direct detection technique [17, 20].

The system is schematically depicted in Fig. 12.5. To achieve the required frequency stability, ultrastable radiation from a low-power seed laser is injected into the transmitter laser. Its output pulse passes a relay optics to get onto the transmitter telescope and out into the atmosphere.

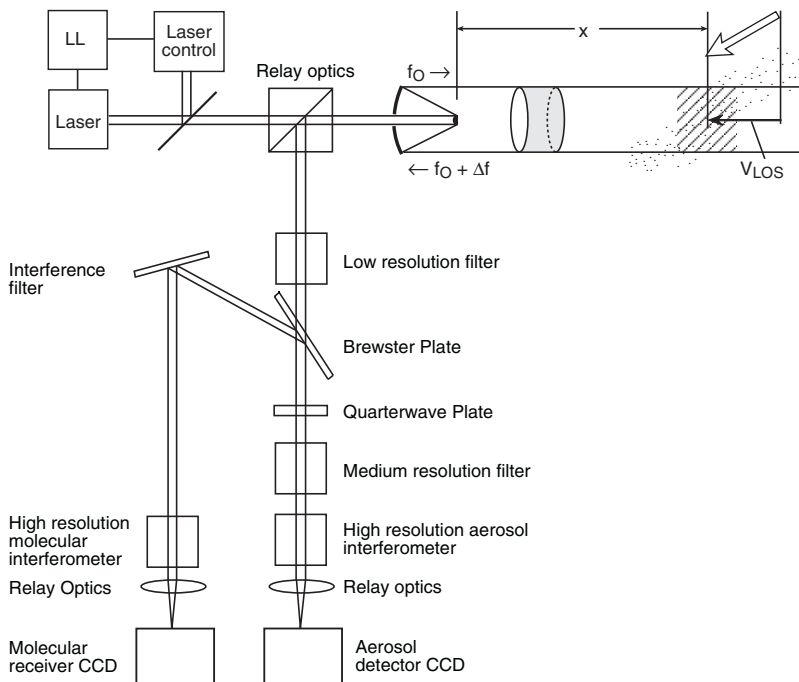


**Fig. 12.5.** Schematic of a direct-detection lidar.

The backscattered radiation is split into two channels, an aerosol and a molecular channel. Both parts pass several filters and are at the end recorded by a CCD detector. A Fabry–Perot and a Fizeau interferometer are used for the molecular and the aerosol component, respectively. This scheme allows one to partially circumvent the problems associated with the superposition of the broad molecule signal with the narrow aerosol peak. To meet eye-safety requirements, the system works with UV radiation around  $0.35\ \mu\text{m}$ .

### Heterodyne Detection

In heterodyne detection the return signal is not passed through one or several narrow-band optical filters. Instead, the return signal is mixed with the radiation from a local optical oscillator (“LO”). The mixed signal contains the sum and the difference frequencies of the two components. The sum is way above the frequency cutoff of the detector, but the difference is a low-frequency signal that can be determined with great accuracy. What is needed for heterodyne-detection lidar is thus a pulsed transmitter laser with high frequency stability of the output frequency  $f_0$  and a second, continuous-wave laser with frequency  $f_{\text{LO}}$ . The mixing results in frequencies  $f_{\text{LO}} \pm (f_0 + \Delta f)$ , where  $f_0 + \Delta f$  is the Doppler-shifted frequency backscattered from the atmosphere. Apart from a DC component, the superposition results in a detector current

$$i_{AC} = \rho \left\{ \sqrt{2P_{\text{LO}}P(x, \lambda)} \cos[2\pi(f_{\text{LO}} - (f_0 + \Delta f))] + \sqrt{2P_{\text{LO}}P(x, \lambda)} \cos[2\pi(f_{\text{LO}} + (f_0 + \Delta f))] \right\}. \quad (12.7)$$

As mentioned, only the first component, or beat signal

$$i_{DET} = \rho \sqrt{2P_{\text{LO}}P(x, \lambda)} \cos[2\pi(f_{\text{LO}} - (f_0 + \Delta f))] \quad (12.8)$$

is measured by the detector, with

|                                 |                                                         |
|---------------------------------|---------------------------------------------------------|
| $\rho$                          | the detector sensitivity,                               |
| $P_{\text{LO}}, f_{\text{LO}}$  | the power and frequency of the reference laser, and     |
| $P(x, \lambda), f_0 + \Delta f$ | the power and frequency of the backscattered radiation. |

The latter two quantities are the only ones that vary with range  $x$ .

The frequency difference between the frequency of the transmitted laser pulse,  $f_0$ , and the local oscillator,  $f_{LO}$ , including the sign of this difference, is determined with great accuracy and maintained as stable as possible during the measurement. It is also a key parameter in the subsequent signal evaluation.

A coherent Doppler lidar (Fig. 12.6) consists in principle of a high-power, frequency-controlled, pulsed laser transmitter (TE), a transmitter-receiver telescope, two heterodyne detectors (D1, D2) in which the local-oscillator radiation is mixed with the outgoing pulse (D1) and with the Doppler-shifted backscatter signal (D2), and a signal processing system (not shown in Fig. 12.6). A locking loop (LL) connects the two lasers. The length of the laser pulse is normally a few microseconds. The temporal distribution of the pulse power is either gaussian (for solid-state lasers) or like a gain-switched spike (for CO<sub>2</sub> lasers). If a CO<sub>2</sub> laser is used at a wavelength around 10.6  $\mu\text{m}$ , a frequency shift  $\Delta f$  of 189 kHz corresponds to a radial velocity component of 1 m/s [21–23].

The optical signal contains speckle which results from constructive and destructive interference of waves scattered by randomly distributed particles. Different shots into the same part of the atmosphere thus lead to different return signals because of the random distribution of the scatterers.

To sum up, heterodyne-detection lidars thus differ from most other lidars by their need for

- a pulsed, narrow-frequency, ultrastable high-power laser,
- a second narrow-frequency laser usually referred to as local oscillator (LO),
- a fast detector in which the return and LO signals are mixed,

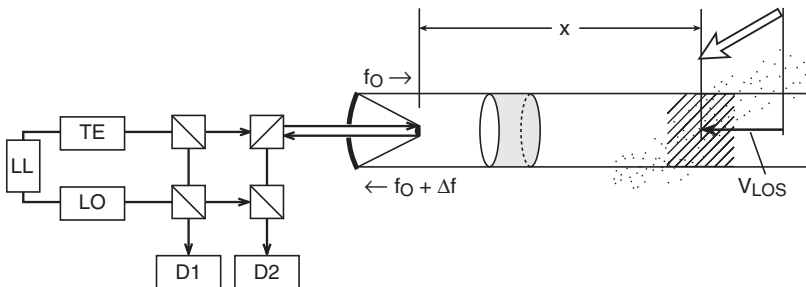


**Fig. 12.6.** Principle of a heterodyne-detection Doppler lidar.

- a second fast detector in which the transmitted and LO signals are mixed (the so-called pulse monitor),
- the time for averaging over several shots to average out speckle, and
- the presence of aerosol particles.

The main assets of the heterodyne-detection technique are the high tolerance of background light and the independence of temperature and all properties of the optical components of the system.

Figure 12.7 shows an example of a lidar heterodyne signal. One can identify a strong signal near the ground, then a weak signal up to a height of 7.3 km where there are few aerosols present, and then a strong signal from a cirrus cloud that extends up to 9.6 km.

### 12.4.3 Scan Techniques

As pulsed Doppler lidars measure profiles of the line-of-sight wind velocity, vertically pointed systems directly provide the profile of the vertical wind velocity. For the horizontal wind, the lidars must be tilted out of the vertical. In this way the horizontal wind produces a line-of-sight component to the lidar signal, and with appropriate scanning schemes the three-dimensional wind vector can be inferred [24, 25]. A necessary assumption is horizontal homogeneity of the wind field over the sensed volume. Vertical homogeneity, however, is not required.

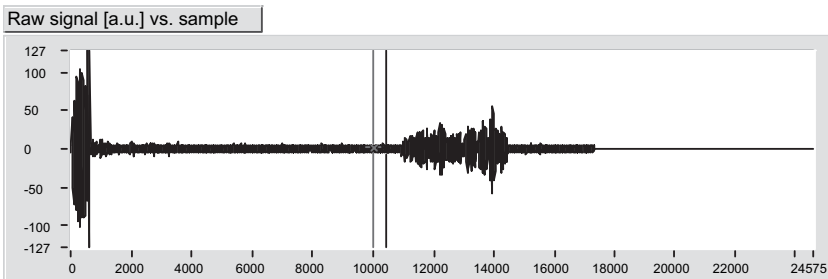


**Fig. 12.7.** Example of a heterodyne lidar signal. The horizontal axis is time-bin (or “sample”) number, i.e., distance, with one sample corresponding to 1.5 m. Because of the paucity of aerosol there is little signal below 7300 m, except for the region close to the ground below about 300 m. The cirrus cloud that begins at 7300 m extends up to 9600 m altitude.

### VAD Technique

When a conical scan is carried out with the apex of the cone at the lidar scanner as depicted in Fig. 12.8 and, for a given height or distance, the velocity signal is displayed as a function of azimuth angle, a plot as the one shown in Fig. 12.9 is obtained. From this display of velocity versus azimuth the technique got its name of velocity-azimuth display, or VAD [26, 27]. In the ideal case of a homogeneous atmosphere the measured LOS component shows a sine-like behavior (Fig. 12.9) given by

$$v_r = -u \sin \theta \cos \varphi - v \cos \theta \cos \varphi - w \sin \varphi, \quad \text{with} \quad (12.9)$$

- $u$  the west–east component,
- $v$  the south–north component,
- $w$  the vertical component,
- $\theta$  the azimuth angle, clockwise from North, and
- $\varphi$  the elevation angle.

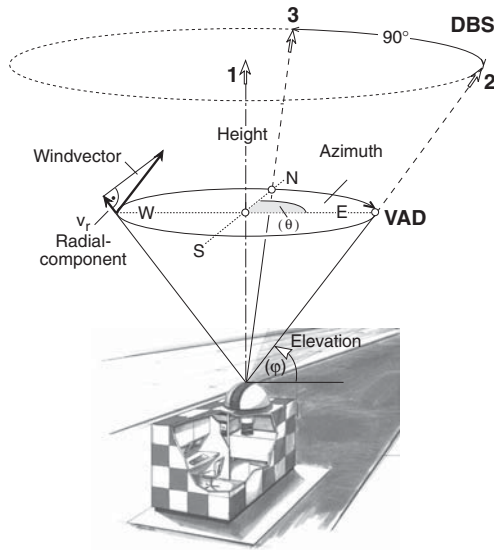


**Fig. 12.8.** Schematic of the scan technique of a Doppler lidar. Lower part: VAD scan, upper part: DBS scan.



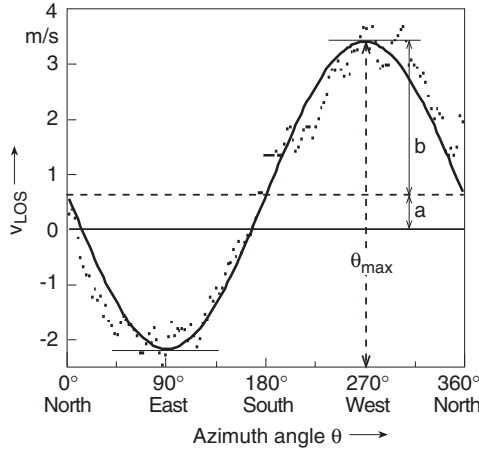


**Fig. 12.9.** Example of sine fitting of the radial wind velocity simulated with the use of the VAD technique.

If we fit this to a function of type

$$v_r = a + b \cos(\theta - \theta_{\max}) \quad (12.10)$$

with offset  $a$ , amplitude  $b$ , and phase shift  $\theta_{\max}$ , we immediately get the three-dimensional wind vector

$$\mathbf{u} = (u, v, w) = (-b \sin \theta_{\max} / \cos \varphi, -b \cos \theta_{\max} / \cos \varphi, -a / \sin \varphi). \quad (12.11)$$

With this, the horizontal wind speed  $u_{\text{hor}}$  is

$$u_{\text{hor}} = (u^2 + v^2)^{1/2} = b / \cos \varphi, \quad (12.12)$$

the horizontal wind direction  $dd$ , as westwind, e.g., blows *from* the west,

$$dd = \theta_{\max}, \quad (12.13)$$

vertical wind velocity  $w$ , defined as positive for wind *up*, is

$$w = -a / \sin \varphi, \quad (12.14)$$

and total wind speed is

$$|\mathbf{u}| = (u^2 + v^2 + w^2)^{1/2}. \quad (12.15)$$

For a VAD scan, a separate sine-wave fit is done for each height interval. From each of those one set of data  $a$ ,  $b$ ,  $\theta_{\max}$  and, consequently,  $u$ ,  $v$ ,  $w$  is obtained for each height interval.

When used with a ground-based system, these formulas can be used as above. For airborne systems, they must be corrected for the movement of the aircraft. Because the speed of the platform is as a rule much higher than the wind speed, the VAD speed is mainly due to the movement of the airplane, and the contribution from the wind results in a small perturbation. To separate the two, the speed and direction of the aircraft must be known with high accuracy.

The smoothness of the sine-wave fit and thus the precision of its parameters depends on instrumental parameters, but also on turbulence and thus on the roughness of the terrain and on weather data such as atmospheric stratification stability. In addition to such lidar data as pulse-repetition frequency and time-bin width, these other factors must be taken into account if a planned measurement is to yield the desired data in a predetermined time. This applies to the VAD and the DBS scan techniques (see below) in the same manner.

### DBS Technique

Under the assumption of cellular flow with little turbulence which would lead to a smooth sinusoidal behavior in the VAD scan, it can be expected that four measurements at azimuth-angle intervals of  $90^\circ$ , or three at  $120^\circ$ , or even two at right angles should be sufficient, along with one measurement in the vertical. For the case of a total of three directions (vertical, tilted east, and tilted north), the three components  $u$ ,  $v$ ,  $w$  are obtained as follows:

$$u = -(v_{r2} - v_{r1} \sin \varphi) / \cos \varphi, \quad (12.16)$$

$$v = -(v_{r3} - v_{r1} \sin \varphi) / \cos \varphi, \text{ and} \quad (12.17)$$

$$w = -v_{r1}. \quad (12.18)$$

Here  $v_{r1}$ ,  $v_{r2}$ , and  $v_{r3}$  are the vertical, east, and north radial velocities, respectively.

This Doppler beam swinging, or DBS, technique is faster and simpler both in the hardware and in the data evaluation algorithm, but lacks the goodness-of-fit information as a measure for the reliability of the results. This shortcoming is partially compensated by information about

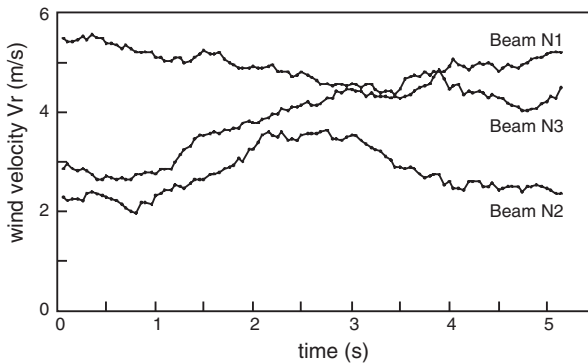


**Fig. 12.10.** Simulation of the behaviour of turbulent wind components in the boundary layer, from Ref. [27].

the temporal behavior of the data. An example is shown in Fig. 12.10 which presents results of a simulation of high-frequency data obtained from three slant lidar beams oriented such that the projections onto the horizontal plane form three angles of  $120^\circ$ . From data such as those of Fig. 12.10 the degree of smoothing (or temporal integration) necessary to obtain the wind speed and direction data required for a given application can directly be inferred. In addition, turbulence is easily determined for any time scale as dictated by the particular process investigated, particularly as turbulence depends critically on ground roughness length and atmospheric stratification stability.

In principle, the VAD and DBS scan techniques can be combined with both direct-detection and heterodyne-detection Doppler wind lidar systems. As parameters such as maximum range, range resolution, temporal resolution (or scan rate), and wind-speed and wind-direction sensitivity all depend on one another and in a somewhat different way for VAD and DBS scans, these dependencies must be known and observed when planning a measurement for a given purpose.

## 12.5 Systems and Applications

There are numerous wind lidar systems in operation and even more in the planning and construction phase today. Only a small fraction of them can be mentioned here. The selection has not been made according to “seniority” or ancientness; instead, we have been trying to show what diverse applications can profit from Doppler wind lidar.

Historically, heterodyne-detection Doppler lidars were the first to offer accurate, dependable results on a routine basis. Direct-detection systems, simpler in design although more critical in components, adjustment and stability, are in a way lagging behind. Following the order in which the two detection schemes have been treated in this chapter, we first briefly describe a direct-detection system built in France and then present different applications of heterodyne-detection lidars, until now the workhorse of Doppler wind lidar. The section concludes with the presentation of a continuous-wave (cw) lidar and another system previously considered as being classified as on the borderline between long-range, depth-resolving lidars and shorter-range non-lidar systems, a laser Doppler velocimeter.

### 12.5.1 Direct-Detection Lidar of the OHP

The Doppler wind lidar of the Observatoire de Haute Provence (OHP) at Saint-Jean-l'Observatoire, France, uses a frequency-doubled, well frequency-stabilized Nd:YAG laser which emits in the green. Laser pulses are sequentially transmitted to three separate telescopes, one oriented vertical, the other two north and east at elevation angles of  $55^\circ$ . The aerosol and molecular scattering components of the return signals are separated using the double-edge technique with a Fabry–Perot interferometer (FPI). Its characteristics have been determined experimentally and least-squares-fitted to the transmission function of a spatially homogeneous FPI. However, the measured calibration curve turned out to be more complex, and the actual transmission function takes into account residual surface inhomogeneities [28, 29]. Figure 12.11 shows the system layout of the OHP direct detection Doppler lidar.

Calibration of the system is carried out when the lidar beam is pointed to the zenith, under conditions of zero wind [29]. When taking measurements, the vertical wind is used as a correction in the evaluation procedure of the horizontal components. Measurement time is typically one minute for each of the three directions. For daylight operation a special procedure is applied to remove the skylight background.

### 12.5.2 Boundary-Layer Flow Measurements with the NOAA Heterodyne Doppler Wind Lidar

Among the many Doppler wind lidars built at the National Oceanic and Atmospheric Administration (NOAA) laboratory at Boulder, CO, one

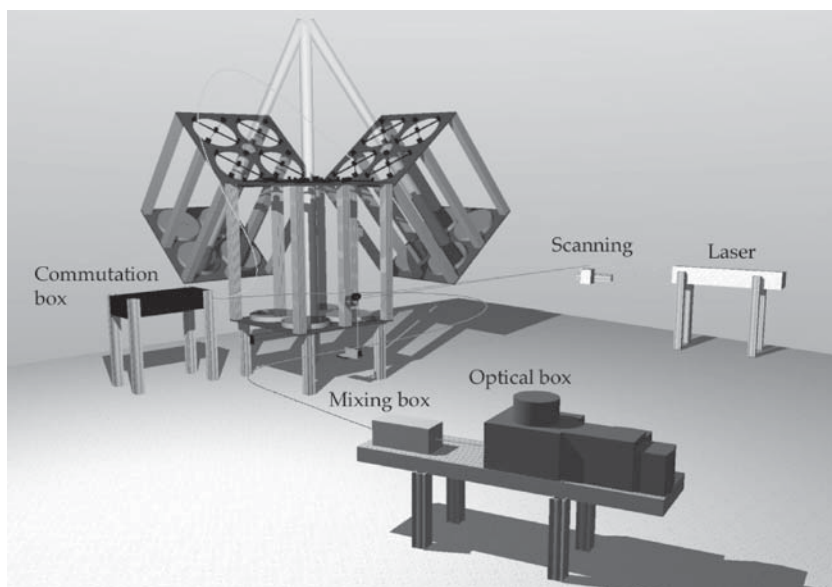


**Fig. 12.11.** OHP direct-detection Doppler lidar design.

of the more recent instruments is the Mini-MOPA system. It is a design based on a  $\text{CO}_2$ -laser Doppler system using a seed laser, or master oscillator (MO), injecting narrow-bandwidth radiation into a second laser, the power amplifier (PA). The system has selectable wavelengths between 9 and 11  $\mu\text{m}$ , 2 mJ output energy, and 300 Hz pulse repetition rate. The maximum range limited by the digitizer is 18 km, with a range resolution that can be selected between 45 and 300 m. The system has been used in many campaigns and is semiautomated, allowing hands-off operation for several hours.

One of the impressive applications of such a Doppler lidar is the representation of the flow in the boundary layer [30]. Figure 12.12 gives an example. This image of the color-coded LOS wind component on an area covering a half-circle of about 8 km radius explains different flow situations from the mountains and back together with the normal wind from the southern basin.

### 12.5.3 Airborne Heterodyne Lidar Within the WIND Project

A relatively recent heterodyne Doppler wind lidar system is the one developed under the Wind INfrared Doppler (WIND) lidar project. It

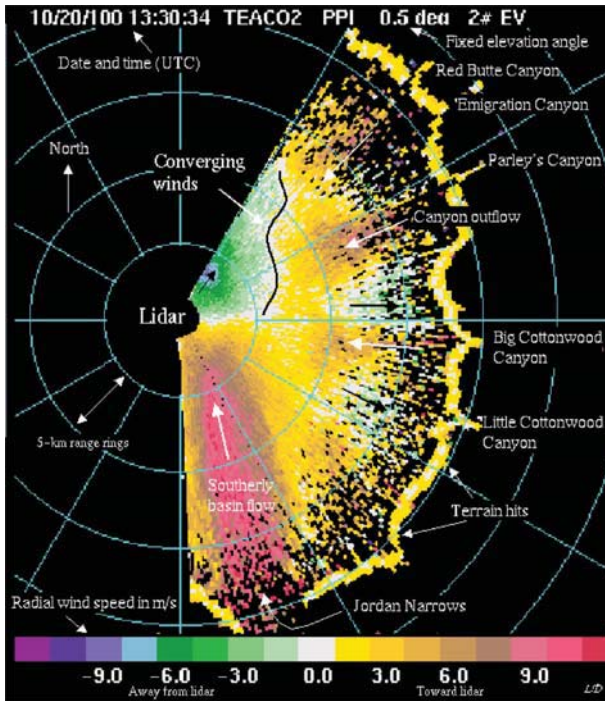


**Fig. 12.12.** Near-horizontal ( $0.5^\circ$ -elevation) azimuthal scan of radial wind velocity [30].

is an airborne system based on the familiar  $\text{CO}_2$  laser concept. The project [31], a French–German cooperation of the Centre National de la Recherche Scientifique (CNRS) and the Centre Nationale d’Etudes Spatiales (CNES) with the Deutsches Zentrum für Luft- und Raumfahrt (DLR), is characterized by two objectives. The first is a significant contribution to mesoscale meteorology by investigation of phenomena like the influence of orography on atmospheric flows, land–sea interaction, the dynamics of convective and stratiform clouds, and the transport of humidity. This defines the requirements on spatial resolution of 250 m in height with a grid size of  $10 \times 10 \text{ km}^2$  and on velocity accuracy of 1 m/s for the horizontal wind component. The second objective is to act as a precursor for the spaceborne global wind measurement system AEOLUS, the Atmospheric Explorer for Observations with Lidar in the Ultraviolet from Space, in the framework of the ESA Atmospheric Dynamics Mission (ADM); such a system is scheduled for launch in 2007 (cf. Chapter 13 of this

book). Airborne Doppler lidars were used in the past [32–35] and are applied also as precursor experiments for spaceborne application of the technique [35].

A validation flight was carried out on 12 October 1999, 13:30 UTC. The wind profiler radar (WPR) [36] of the Meteorological Observatory Lindenberg (MOL) of the German Weather Service DWD was used as the reference instrument, extrapolation to more remote areas was done with the local model (LM) of the DWD. This model has a horizontal resolution of 7 km. It is a non-hydrostatic model with 35 altitude levels and includes turbulent vertical exchange. Airborne wind lidar systems are expected to meet at least the accuracy and horizontal-resolution specifications of state-of-the-art numerical models [37].

Figure 12.13 shows the result of the wind profile determination. Within the statistical variations, the agreement is perfect.

The WIND instrument is a flexible and modular system for airborne measurements of mesoscale wind phenomena. It provides actually an accuracy of 1 m/s for the horizontal wind in a volume of the size of

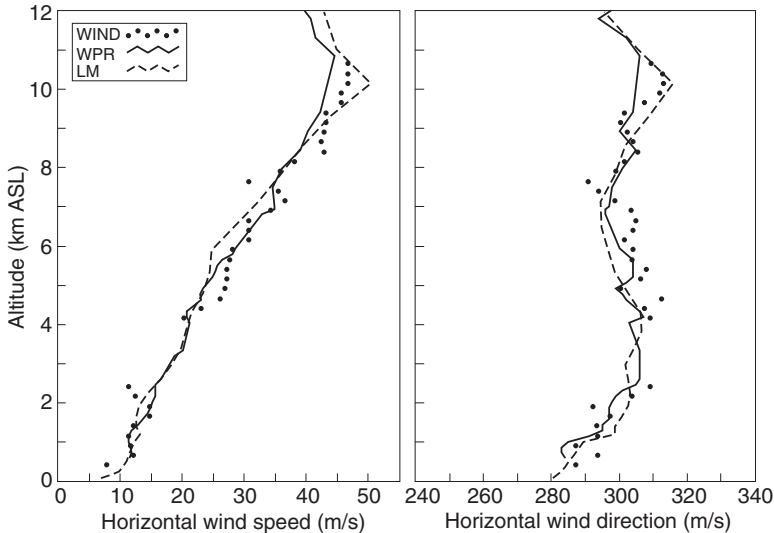


**Fig. 12.13.** Comparison of wind profiles determined with the WIND instrument (dotted lines), the radar wind profiler (WPR, solid lines), and the WPR data extrapolated to the area covered by the WIND system (dashed line). WIND data comprise 5 conical scans, or  $>100$  s of measurement time, in a  $10 \times 50$  km<sup>2</sup>-size horizontal grid. WPR data are averaged over 30 minutes. Note the suppression of zero in the wind-direction scale.

10 km  $\times$  50 km  $\times$  250 m in the boundary layer. An improvement of the resolution to 10 km  $\times$  10 km  $\times$  250 m appears realistic as a near-future goal. Other uses in the scanning or non-scanning mode are also possible, e.g., the determination of single line-of-sight wind profiles to simulate the performance of spaceborne Doppler lidars.

#### 12.5.4 Ground-Based Continuous-Wave Heterodyne Lidar for the Measurement of Wake Vortices

The measurement principle for determining aircraft wake vortices with a Doppler wind lidar is illustrated in Fig. 12.14. These inhomogeneities of the wind-field vector can be dangerous to aircraft, especially in landing operations. Every plane, when in flight, generates in its wake a pair of counterrotating horizontal vortices. When planes fly behind one another in close succession, wake vortices can present a considerable hazard. At present the U.S. Federal Aviation Administration (FAA) mandates minimum distances for aircraft during instrument landing conditions. These differ with airplane size. There are three categories: heavy, large, and small.

These wake vortices are invisible to the eye, but can be detected with Doppler wind lidar. The lidar scans the air volume in a vertical plane perpendicular to the landing runway. The radial velocity can reach values around 20 m/s, depending on the type of aircraft, its weight and its takeoff velocity. It can be used to calculate the rotational velocity of the vortex. The scheme of the measurement is sketched in Fig. 12.14. The change in  $v_{\text{LOS}}$  caused by the vortex, divided by the cosine of the

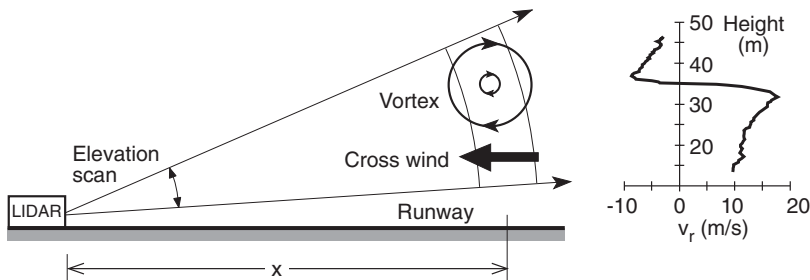


**Fig. 12.14.** Left, measurement principle for determining aircraft wake vortices with a Doppler wind lidar.  $v_r$  radial or line-of-sight wind velocity. Right, vertical distribution of crosswind at distance  $x$  (which might be the position of the landing-runway centerline).



lidar elevation angle, directly yields the amount and spatial distribution of excess cross wind for the landing airplane [38, 39].

Continuous-wave CO<sub>2</sub>-laser Doppler lidars like the one used here are in operation at the French Office National d’Études et de Recherches Aéronautiques (ONERA), the Deutsches Zentrum für Luft und Raumfahrt (DLR), and the Massachusetts Institute of Technology (MIT) in the United States. The European Community has started a program to both detect and forecast wake vortices. For a better range resolution pulsed systems (2-μm laser Doppler system from Coherent Technologies (CTI, <http://www.ctilidar.com>)) are used [40]. The parameters of such a lidar are listed in Table 12.1.

Clearly, large-aircraft wake vortices are not stationary. Their displacement can easily be followed with lidars. Figure 12.15 shows an example of the determined position of the core of the vortex.

12.5.5 Clear-Air Turbulence

A key application of Doppler wind lidar to aircraft safety is also the measurement of clear-air turbulence (CAT), a hazard hard to determine with any other means. Considerable progress has been made in this important field in recent years. Figure 12.16 shows a time-series plot of the velocity estimates, along with *in situ* true-airspeed (TAS) measurements. As can

Table 12.1. Main parameters of the 2-μm pulsed Doppler lidar

|                     |                                        |             |
|---------------------|----------------------------------------|-------------|
| Slave laser:        | Type                                   | Tm:LuAG     |
|                     | Wavelength                             | 2022.54 nm  |
|                     | Pulse energy                           | 2.0 mJ      |
|                     | Pulse length (FWHM)                    | 400 ± 40 ns |
|                     | Pulse repetition rate                  | 500 Hz      |
|                     | LO/SO frequency offset                 | 102 ± 3 MHz |
| Telescope:          | Type                                   | off-axis    |
|                     | Aperture                               | 108 mm      |
| Scanner:            | Oscillating mirror vertical scan range | ≥ 20°       |
|                     | Scan duration                          | ≈ 11 s      |
|                     | Fly-back time                          | ≈ 0.5 s     |
| Data acquisition:   | Concept of early digitizing            |             |
|                     | Sampling rate                          | 500 MHz     |
|                     | Sample length                          | 0.3 m       |
| Measurement range:  | Processed data                         | 500–1100 m  |
| Spatial resolution: | Along LOS                              | 3 m         |
|                     | Perpendicular to LOS                   | 0.9–1.9 m   |

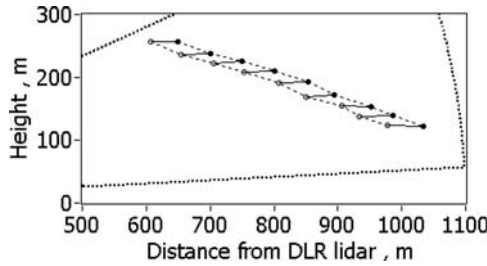


**Fig. 12.15.** Wake vortices of large aircraft measured on 13 June 2002 with the DLR wake vortex lidar. Trajectories of port vortex (open circles) and starboard vortex (full circles) observed during 9 consecutive scans.

be seen, due to CAT the radial velocities at different ranges in front of the aircraft vary by  $>20$  m/s or  $>70$  km/h within 3 minutes.

### 12.5.6 Remote Wind Speed Measurements for Wind Power Stations

Installations that also critically depend on wind speed and direction are wind power stations. If blade pitch and rotor orientation are properly adapted to the prevailing wind, then both efficiency and safety of the facility can be maximized. However, the feedback mechanism of the

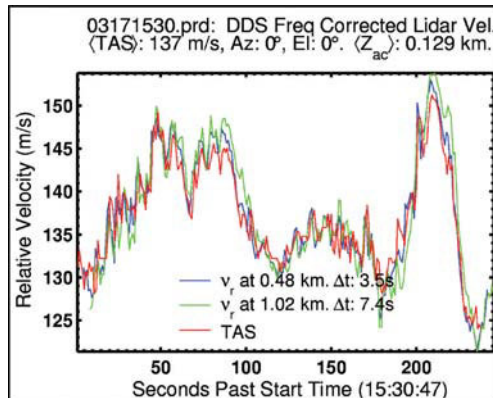


**Fig. 12.16.** Velocity versus time for an airborne clear-air turbulence Doppler lidar [40]. The three traces represent the lidar measurement 3.5 s or 0.48 km (blue) and 7.4 s or 1.02 km (green) ahead of the time of passage, as a function of distance, and the result of the *in situ* (true airspeed, TAS) measurement (red trace).

systems is too slow to adequately follow changes in wind speed and direction. It is therefore necessary to measure these quantities at a distance of roughly 150 m windward of the turbine.

It may seem as if a lidar was needed for the purpose. However, this is not the case. A normal cw laser Doppler velocimeter (cf. Section 3.3) will be perfectly sufficient for the purpose. A corresponding system for installation at the top of the turbine is currently under development [41]. First tests with a CO<sub>2</sub>-laser source have shown encouraging results.

## 12.6 Future Developments

Like most other lidar developments, wind lidar is also a field in which instruments are continuously improved, reduced in size, weight, power consumption, and cost. Instruments are getting more highly integrated to make them easier to align, adjust and operate. Simultaneously and partially caused by this instrument improvement process, the number of fields in which wind lidar systems are being used increases. The development [42] as well as the new uses are greatly facilitated if a certain standardization takes place. In this section one example is given for each of these tendencies to show in which direction near-future work in wind lidar is likely to go.

### 12.6.1 Instruments

In the field of instruments, work toward spaceborne systems is of course one of the important items (cf. Chapter 13 of this book). This development started as soon as the end of the 1980s [43, 44] and is in full swing today. What is new, however, is an “instrument” that is sheer software and which we call a virtual instrument. Virtual instruments [45] represent powerful tools to test and investigate system performance in various combinations of components and under varying atmospheric conditions without any hardware development. The data sets generated with such virtual instruments can be used to submit components to further tests, to try out new ones, to improve and validate program modules like signal processors, and to carry out virtual experiments.

In the DLR virtual Doppler lidar (see Fig. 12.17) the parameters that can be varied include laser wavelength, pulse power, and transceiver characteristics. The pulse shape can be either gaussian (as for solid-state lasers) or spiked (as for CO<sub>2</sub> lasers). Detection can be direct or of the

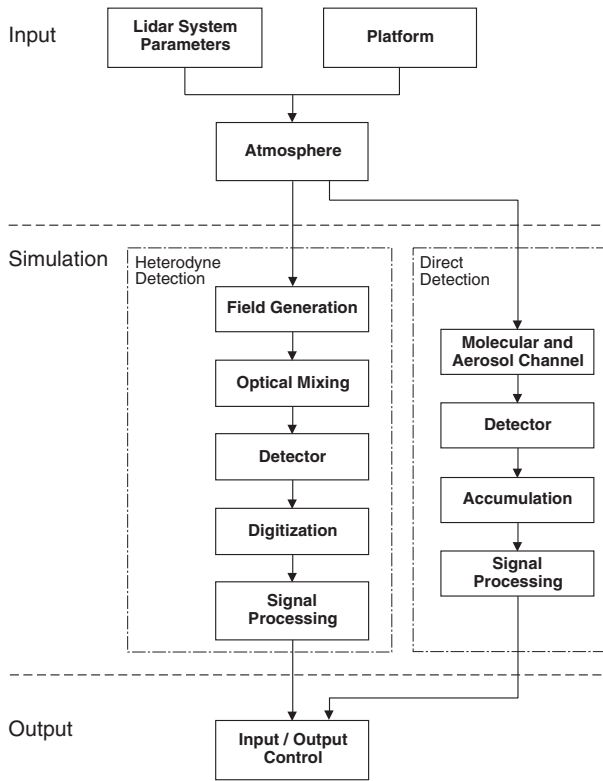


**Fig. 12.17.** Block diagram of the DLR virtual instrument.

heterodyne type. The platform can be chosen to be an aircraft, satellite, or ground-based station, all with characteristic parameters. For the virtual lidar the atmosphere is “sliced” into height intervals of 1.5 m minimum thickness. In the slices the optical beam is scattered and absorbed with uniform coefficients  $\beta$  and  $\alpha$ . Clouds strongly affect the values of both  $\beta$  and  $\alpha$ . Noise is generated with a module called AGNA, for Additive Gaussian Noise Approximation; AGNA was originally developed at the Technical University (TU) Vienna [46]. In the digitization and signal-processing modules a number of options is available including a variant in which return signals are treated pair by pair (pulse-pair or PP mode), and one that observes the criterion of maximum likelihood (ML mode). The direct-detection virtual instrument can work on two techniques, the double-edge (for the exploitation of the molecular signal) and the multichannel-Fizeau technique (for the aerosol signal). The system

is written in LabVIEW<sup>®</sup>. Its result is the comparison of a calculated (“measured”) wind profile with the input, or “true,” profile.

There are other virtual instruments as well. One of them is based upon the Delphi Study of ESA [46] and the improved ALIENS simulator [47]. By adding scanning capability and the possibility to operate aboard a moving platform, a new dynamic version of a virtual instrument is obtained. For “experiments” it requires a three-dimensional model of the atmosphere.

### **12.6.2 Weather Forecast**

A field that heavily relies on the availability of high-accuracy, high-resolution wind data is meteorology. The lack of global wind data is indeed one of the major deficiencies of the current meteorological network. In fact, one of the hot candidates to provide a great deal of the necessary wind parameters is a future spaceborne Doppler lidar [48, 49]. To determine exactly what is needed and which configuration will provide the most reliable results can no better be determined than with one of the virtual lidar instruments described above. A study is actually under way to estimate the effect of lidar winds on numeric weather prediction [50].

### **12.6.3 Standardization**

Work that is done independently in different places necessarily creates different technical terms for the same objects and notions. Different parameter values are chosen as the basis for key performance characteristics. Different procedures are followed when a measurement or measurement campaign is planned, prepared and carried out and when the data are processed, evaluated and presented. Often certain of those procedures proved technically superior to others. To make the corresponding knowledge publicly available, guidelines proved very useful. Not only do these recommendations help the manufacturers, they also allow users to decide whether or not the lidar technique can meet their goal, to clarify their expectations and to specify their requirements on a lidar for a given purpose. The Doppler wind guideline of the German Commission on Air Pollution Prevention [42] was one of the first to appear in a series of guidelines on quantitative remote sensing techniques and served as a kind of model to the other ones that followed [51].

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# 13

## Airborne and Spaceborne Lidar

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### 13.1 Introduction

The evolution of lidar, from those early ground-based measurements to our first long-duration spaceborne experiments, is schematically represented in Fig. 13.1. It depicts the first lidars in the 1960s as ground-based, followed by systems first flown in 1969 on small aircraft, and followed in the late 1970s by lidars flown on large aircraft capable of long-range measurements. Starting in 1979, flights aboard high-altitude aircraft were accomplished where data were taken at approximately 20 km altitude. The depiction finishes with the first spaceborne lidar using Shuttle for the 11-day flight of LITE, the Lidar In-space Technology Experiment in 1994, and finally, the first long-duration spaceborne low-Earth-orbit flight, that of the Geoscience Laser Altimeter System (GLAS) launched aboard ICESat in January 2003.

The above were pathfinders in lidar's evolution, with the first flights utilizing elastic backscatter for cloud and aerosol measurements. This chapter will describe the airborne and spaceborne firsts, including flights of lidars using other techniques like DIAL, and then present a more detailed description of specific examples of both airborne and spaceborne missions. It will conclude with a look into the future. It will not cover in any detail airborne or spaceborne laser altimeters, bathymeters or Doppler lidars.

### 13.2 History of Airborne Lidar

After early successes in the 1960s and early 1970s, using lidars to probe the atmosphere, researchers began building systems to be carried aboard



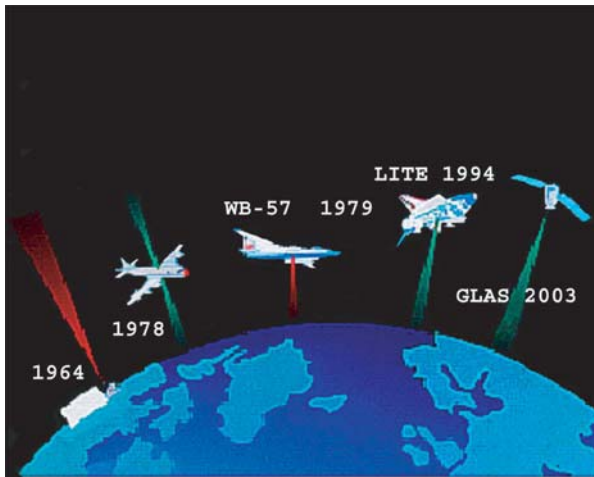


**Fig. 13.1.** An artist sketch depicting the evolution of lidar “firsts.”

aircraft. These were attractive in order to obtain a more “regional” capability, to have the ability to move to the area of concern for the measurements needed, or to capture better the phenomenon of interest. The first airborne lidar measurements occurred in 1967, when S. Harvey Melfi flew aboard a NASA Langley Research Center (LaRC) T-33 aircraft over Williamsburg, VA, at constant altitudes making measurements with a forward-looking, very modest lidar, while the author made “uplooking” simultaneous measurements with a ground-based zenith-pointed lidar [1]. The objectives of this airborne research were to develop lidars for the detection of clear air turbulence [2]. The first “downlooking” airborne lidar was built by the Stanford Research Institute and flown in 1969 for making lower tropospheric aerosol measurements during the Barbados Oceanographic and Meteorological Experiment [3]. The first “uplooking” airborne lidar was a two-wavelength elastic backscatter system built by LaRC that made aerosol profile measurements to validate the satellite Earth-orbiting mission called the Stratospheric Aerosol Measurement-II (SAM II) launched in October 1978 aboard the Nimbus-7 spacecraft [4]. This “ground truth” experiment was staged out of Sondrestrom, Greenland, in November 1978. It was followed in July 1979 by similar flights staged out of Poker Flat, Alaska. The Poker Flat mission was directed at validating both SAM II and another satellite mission called the Stratospheric Aerosol and Gas Experiment (SAGE)

that was launched in February 1979 aboard the Applications Explorer Mission satellite [5]. Whereas SAM II measured stratospheric aerosols in the polar regions, SAGE made its measurements on a much more global basis of not only aerosols but also ozone. The aircraft measurements of aerosol backscatter were made to coincide with the satellite measurements in time and space. Simultaneous balloon-borne and aircraft aerosol *in situ* measurements were also timed to occur during satellite overpasses. The validation measurements at Poker Flat included balloon-borne and rocket-borne measurements for validating the SAGE ozone measurements.

Although not the topic of this book, there were laser altimeters and lidar bathymeters that flew aboard aircraft in the mid-1970s also. For example, in 1974 a NASA C-54 aircraft flew a N<sub>2</sub>-Ne laser over the Chesapeake Bay and around Key West, Florida. It flew at low altitudes and measured water depths in clear water [6].

The first lidar built for a high-altitude aircraft was the Cloud Lidar System (CLS) built by NASA Goddard Space Flight Center (GSFC) and flown aboard the WB-57 aircraft in 1979. By necessity, it had to operate autonomously with minimum pilot interaction [7]. The experience gained would be used for future spaceborne lidar, including simulations needed for designing a spaceborne lidar.

The aerosol and cloud lidars continued their measurement campaigns aboard aircraft into the 1980s, 1990s, and to the present, improving their capabilities as new technologies became available and as new applications were needed. Multiple wavelengths and polarization measurement techniques were incorporated. Higher repetition rates and more efficient lasers, as well as improved and faster data capture and storage devices, helped expand airborne lidar applications. The airborne aerosol and cloud lidars circled the globe mapping stratospheric volcanic layers, Saharan dust, stratospheric aerosols, and polar stratospheric clouds (PSC)s, for example. In addition, other lidar measurement techniques began to be used aboard aircraft. As early as 1980, NASA LaRC operated the first airborne UV DIAL system for ozone measurements in a “down-looking mode” [8]. Shortly thereafter, an airborne water vapor DIAL system was first developed and flown by NASA LaRC in 1982 [9]. It too was operated in a “down-looking” mode. Atmospheric pressure was first measured in 1985 with a down-looking airborne DIAL system, built by NASA GSFC using a tunable alexandrite laser [10]. The LaRC DIAL system was re-configured to make the first “up-looking” DIAL measurements of ozone because a relatively high-resolution ozone measurement

was needed for the Antarctic ozone hole campaign that was staged out of Punta Arenas, Chile, in 1987. Staging out of Punta Arenas allowed data to be taken inside the Antarctic vortex [11]. These measurements, along with a number of other types of measurements to characterize ozone photochemistry, were essential for understanding how the ozone hole formed.

An airborne system using resonance fluorescence to measure Na in the mesosphere was first flown in 1983. In this way, mesopause density perturbations were investigated [12].

The first DIAL measurements aboard a high-altitude aircraft, in this case an ER2 aircraft, were made with a system called LASE (Lidar Atmospheric Sensing Experiment), which was built for water vapor profiling throughout the troposphere [13]. In addition to allowing measurements of water vapor over large ground distances during flights of the ER2, it also represented a test bed for future spaceborne DIAL systems, in this case for applications using DIAL.

### **13.3 History of Spaceborne Lidars**

It became clear during the early atmospheric applications of lidars that a spaceborne lidar orbiting Earth would yield an enormous science payoff. It wasn't surprising then that in the 1970s and 1980s NASA and ESRO (later ESA) put together groups to study the capabilities of lidar on satellite platforms. Because of the heavy weight and high power requirements for those early lidars, the obvious platforms for demonstrating lidar capabilities were Spacelab and Shuttle.

Specific proposals for building and flying a spaceborne lidar have usually been tied to a particular space initiative like NASA's Space-lab and Shuttle programs, or the cooperative Earth Observing System (EOS) sponsored by the European Space Agency (ESA), NASA, and Japan's NASDA. Technical feasibility studies have been carried out for these programs by groups of scientists primarily engaged in lidar and/or atmospheric research. Examples of these studies include the Atmospheric, Magnetospheric and Plasmas in Space (AMPS) payload for Spacelab/Shuttle [14], Atmospheric Research using Spacelab-borne Lasers [15], the Shuttle Atmospheric Lidar Research Program [16], the ESA Space Laser Applications and Technology (SPLAT) Workshop [17], the Lidar Atmospheric Sounder and Altimeter (LASA) Instrument Panel

Report [18], and the LASER Atmospheric Wind Sounder (LAWS) Instrument Panel Report [19]. A number of subsequent studies followed like the German ALEXIS Phase A study [20], and ESA studies of Laser Sounding from Space [21] and the Atmospheric Laser Doppler Instrument [22]. The Proceedings of International Laser Radar Conferences (ILRCs) as well as the archival journals are also rich sources of information on the use of lidars from space to measure atmospheric composition and structures. These studies and papers have shown, with varying degrees of resolution and sophistication, the feasibility of spaceborne lidars to measure the height of the planetary boundary layer, the vertical distribution of aerosols, clouds and trace gases such as ozone and water vapor, tropospheric winds and the vertical distributions of atmospheric pressure and temperature. Lidars provide a new measurement capability for many of these parameters, increased vertical resolution for others, and a daytime capability for some. All of these measurements can be accomplished to some degree with today's technology.

The EOS was established to understand the fundamental, global-scale processes that govern the Earth's environment. The program was to include a series of polar and low-inclination platforms to be flown beginning in the late 1990s and into the first decade of 2000. Proposals were solicited for two generic EOS lidar facilities: LASA for a U.S. platform, and ATLID (Atmospheric Lidar) for an ESA platform. Proposals for membership on the LAWS research facility instrument team for the Japanese platform were also solicited. A number of very interesting and well-conceived proposals were received. NASA, however, decided to postpone the implementation of LASA. Phase B study approval was given to the LAWS activity, but it was subsequently deselected in 1994. Phase B studies for the ESA facility were also conducted but approval to continue was not implemented. Similarly, NASDA was developing the Experimental Lidar in Space Experiment (ELISE) for flight aboard the Mission Demonstration Satellite 2 (MDS-2). The MSD-2 flight was cancelled, however, because of the launch failure of the H-II rocket in November 1999 [23, 24]. Therefore, there were no long-duration flights approved or flown during the late 20th century. However, the modification of the EOS research facility instrument called GLAS (Geoscience Laser Altimeter System) was studied with the intent to perform some atmospheric measurements such as cloud top and height of the planetary boundary layer (PBL) determinations during its approved ice altimetry mission. And, in addition, ESA decided to proceed with spaceborne

lidars in the first decade of the 21st century as will be described later in this chapter.

### 13.4 The Use of Airborne Lidar

Airborne lidars have been used for a number of atmospheric applications. These include studies of the planetary boundary layer, long-range transport of pollutants, power plant plumes, volcanic aerosols in the stratosphere, mesospheric and stratospheric winds and gravity waves, Saharan dust, PSCs, water vapor and the hydrologic cycle, ozone associated with biomass burning, Arctic and Antarctic ozone associated with the ozone hole and ozone depletion, and the validation of satellite experiment measurements in the stratosphere and troposphere. Some of these lidar measurements and studies have been part of a larger campaign or intensive field study. Airborne lidars permit measurements over regional areas with the same lidar in downlooking or uplooking configurations, and over shorter times than possible using a ground-based or ocean-based mobile system. They allow one to measure in areas not easy, or in some cases, not practical, to access. Missions to the polar regions for measuring PSCs and/or ozone are good examples of this unique capability. Airborne systems can also fly above weather systems to ensure measurements can be made. Because they can fly faster than air mass movements, they can measure large-scale patterns. For down-looking systems, the inverse range-squared ( $1/r^2$ ) decrease in backscattered signal is somewhat compensated for by the increased backscattering due to increased atmospheric density with increasing range. This is realized for spaceborne lidar too. An up-looking lidar, as well as a ground-based lidar, has the disadvantage of the  $1/r^2$  decrease in signal between near- and far-field measurements that require a very large dynamic range. The disadvantages of airborne lidars include the paucity of flight opportunities, the inherent costs for aircraft operation, and the somewhat increased system complexities due to space, window, and power constraints. This is especially true for high-altitude aircraft like the ER-2. Aircraft platforms also severely restrict the telescope receiver size that can be accommodated, and can increase the AC frequency, vibration, temperature, voltage, and G-force variations experienced. The issue of eye safety is exacerbated in the case of an airborne lidar and must be carefully dealt with for all airborne flights. Reducing power, increasing altitude, and

changing output wavelength are all used to ensure an eye-safe level for observers on the ground or in lower or higher flying aircraft.

### 13.4.1 Elastic Backscatter

As stated in the history section of this chapter, the first airborne lidar measurements were of aerosols and clouds by down-looking and up-looking lidars. Aerosols play an important role in visibility reduction, pollution, cloud formation and lifetime, atmospheric chemistry, and climate forcing. Information on aerosols is gained by multiwavelength measurements (size) and depolarization measurements (shape). Raman lidar can yield information on aerosol backscatter and aerosol extinction by measuring both elastically and inelastically backscattered radiation (cf. Chapter 9 of this book). Aerosol and molecular backscattering can also be separated using very-high-resolution lidar (HSRL, Chapter 5). The first airborne aerosol measurements mentioned earlier were made with a down-looking lidar aboard an Air Force 130B aircraft over the Caribbean in late June through early July 1969 using a Nd:YAG laser operating at 1 pulse per 3.5 seconds. About 5000 profiles were made of aerosols beneath clouds including what was thought to be Saharan dust [3]. Other airborne measurements followed in the 1970s and early 1980s as systems were explicitly built for tropospheric aerosol measurements [25–29]. The 1977 flights of an Airborne Science Spacelab Experiments System Simulation (ASSESS) aboard the NASA CV990 aircraft were particularly interesting in that it was built to be a proxy for a future spacelab experiment. “Payload specialists” (PS) were trained to operate the lidar. The lidar incorporated two Nd:glass lasers so that the PS could switch from one laser to the other, and even change flashlamps. A 10-day flight series was carried out during May 1977 [30].

In 1978, the first up-looking airborne lidar (Fig. 13.2) was flown on the NASA P-3 aircraft to Sondrestrom, Greenland where it was the centerpiece of the SAM II ground truth campaign [5, 31, 32]. This airborne system flew along tracks between the sun and satellite in order to make aerosol profile measurements as close as possible to those made by the SAM II solar occultation measurements. This lidar was subsequently used in a number of satellite validation campaigns for SAGE and SAGE II [32], for studies of the polar vortex [33], the impact of volcanic eruptions on stratospheric aerosols such as those from Mt. St. Helens [34], El Chichon [35], and Pinatubo [36]; and for



**Fig. 13.2.** LaRC aircraft Lidar (up-looking) aboard the NASA P-3 in 1982 during an El Chichon survey mission.

PSC studies [37–41]. This system has provided unique and important data for validating these spacecraft instruments and understanding the observed phenomena.

Many down-looking lidars have been built and flown after the original ones in 1969 through the early 1980s. The NASA Global Tropospheric Experiment (GTE) included many deployments of an airborne lidar built at NASA LaRC [8, 9, 42, 43]. Biomass smoke and other aerosols were studied over the Atlantic Ocean, western and northern Pacific, and the Canadian boreal forest. Many other examples can be found in the literature including in the proceedings of the biennial International Laser Radar Conferences (ILRCs.) The Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX) conducted off the east coast of the U.S. near 38°N was a recent example of the application of airborne lidars in a major aerosol climate forcing campaign [44, 45]. Other examples include the Global Backscatter Experiment (GLOBE) Pacific missions in 1989 and 1990 [46], the Pacific 1993 experiments in Vancouver, British Columbia [47], the Indian Ocean Experiment (INDOEX '99) as described in Pelon et al., 2001 [48], the study of Saharan dust [49] and aerosols over the Atlantic Ocean [50].

High-altitude aircraft like the WB-57 and ER-2 have provided platforms for lidars to study aerosols and clouds [7, 51]. Cloud measurements by lidar are similar to aerosol measurements except clouds are made up of much larger particles and attenuation through clouds is higher. Lidars

are particularly well suited for the study of optically thin cirrus and other clouds and, of course, cloud top or bottom heights. Multiple scattering occurs in optically thick clouds and affects their measurements and apparent thickness.

### 13.4.2 Resonance Fluorescence

In 1983, the first airborne resonance fluorescence Na lidar was flown to show the feasibility of this technique [12]. The University of Illinois group under the direction of Professor Gardner pioneered the use of aircraft to extend lidar observations over long baselines to study middle atmosphere temperatures and to study the horizontal wavenumber spectra of gravity-wave-induced density perturbations. They conducted major campaigns in the equatorial Pacific [1990 and 1993 Airborne Lidar and Observations of the Hawaiian Airglow (ALOHA) Campaigns] and Canadian Arctic (1993 Arctic Noctilucent Cloud Campaign). This work is described in several special issues of *Geophysical Research Letters* [52, 53] and the *Journal of Geophysical Research, Atmospheres* [54]. In November 1998, the group flew an iron (Fe) atom density lidar to Okinawa, Japan to study meteor ablation during the Leonid meteor shower [55]. In the summer of 1991, during the Arctic Mesopause Temperature Study, the group flew a Rayleigh/Fe Boltzmann Temperature Lidar to the North Pole where they were the first to measure upper mesosphere and lower thermosphere temperatures, mesopause region Fe densities, and polar mesospheric clouds (PMCs) over the geographic and geomagnetic North Poles [56, 57]. Figure 13.3 is a photograph of the Rayleigh/Fe Boltzmann temperature lidar in operation aboard the SF/NCAR Electra aircraft.

### 13.4.3 Raman Scattering

A Raman lidar system was developed by NASA GSFC [58] to measure methane ( $\text{CH}_4$ ) as a conserved tracer for polar ozone determinations in the lower stratosphere. The system demonstrated its capability to measure water vapor ( $\text{H}_2\text{O}$ ) in the lower stratosphere and for profiling temperature from above the aircraft to about 70 km [59]. Another lidar system was developed by NASA GSFC using this technique and molecular backscatter to derive atmospheric density and temperature from a few kilometers above the aircraft to approximately 40 km altitude for flights during the SAGE Ozone Loss and Validation Experiment





**Fig. 13.3.** Dr. Xinzchao Chu from the University of Illinois at Urbana-Champaign operating the Rayleigh/Fe Boltzmann temperature lidar aboard the NSF/NCAR Electra aircraft, 1991. (Courtesy of C. Gardner.)

(SOLVE). The NASA LaRC 532 and 1064 nm backscatter aerosol lidar was combined with the GSFC system, along with a GSFC DIAL lidar using a xenon chloride (XeCl) laser for  $O_3$  measurements. The combined instrument is called the Airborne Raman Ozone, Aerosol and Temperature Lidar (AROTEL). It was flown during the December 1999 to March 2000 SOLVE-1 campaign [60–62] and during the December 2002 to February 2003 SOLVE-2 campaign.

#### 13.4.4 Differential Absorption

The first airborne UV DIAL system for ozone profiling was flown in 1980. It made ozone and aerosol measurements in a down-looking

mode as part of the Environmental Protection Agency's (EPA) Persistent Elevated Pollution Episodes (PEPE) field experiment conducted over the east coast of the U.S. [8, 9]. This system has evolved over the years into the system shown in Fig. 13.4 capable of making uplooking and downlooking profile measurements [63]. For ozone measurements it uses two frequency-doubled Nd:YAG lasers operating at 30 Hz to sequentially pump two dye lasers that are frequency-doubled into the UV to produce the on-absorption line 288.2 nm (for troposphere) or 301 nm (for stratosphere) outputs, along with the off-absorption line outputs at 299.6 nm or 310 nm. The residual 590 to 620 and 1064 nm wavelength outputs are used for aerosol and cloud measurements. The data are processed in real time onboard so that they can be used for mission planning or for diagnostics of the system, and to ensure high-quality data capture. Airborne DIAL systems were developed by other groups for various applications during this period also [64–69]. The first water-vapor DIAL system was flown in 1982 by NASA LaRC [8, 70]. Similar to ozone DIAL, this system evolved [71], and a number of water-vapor DIAL systems were developed and flown by other groups [72–77].

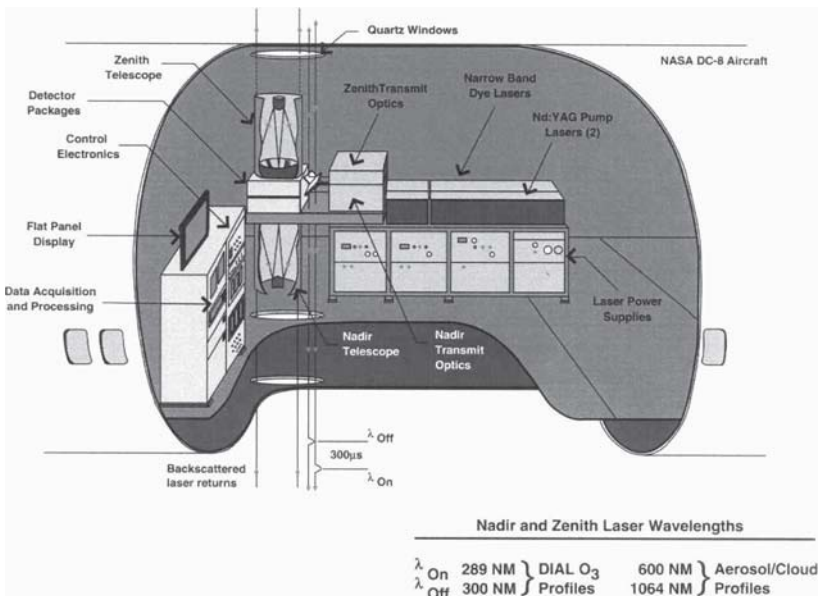


**Fig. 13.4.** Artist's sketch of the NASA LaRC DIAL system used during the Airborne Arctic Stratospheric Expedition. (Courtesy of E.V. Browell.)

As inferred earlier, the NASA LaRC UV DIAL system has been involved in an exceedingly large number of field experiments. Figure 13.5 depicts that record on a world map. It has flown over the last 22 years in 23 major NASA field experiments with 18 being international. Parameters for their system are given in table 13.1. A summary of this work can be found in Ref. [78]. Figure 13.6 presents an example of simultaneous tropospheric ozone and aerosol measurements taken with the LaRC DIAL system.

The first DIAL measurements from a high-altitude aircraft were made by LASE aboard the NASA ER-2 using a titanium:sapphire ( $\text{Ti:Al}_2\text{O}_3$ ) laser emitting at 813–818 nm, and was developed as a step toward a space-based  $\text{H}_2\text{O}$  DIAL system [13, 79]. The pilot could switch absorption lines for different altitude sensitivity. Ten LASE flights were made between September 8 and 26, 1994, during the LITE mission for a total of 60 flight hours on the ER-2. Figure 13.7 shows an example of LASE  $\text{H}_2\text{O}$  and aerosol data taken from aboard the NASA ER-2 aircraft. Through a validation program, LASE was found to agree with other measurements to an accuracy of 6% or 0.01 g/kg, whichever is greater, over the troposphere [80]. Subsequently, LASA on the ER-2 has been involved in the 1996 TARFOX field experiment conducted off the coast of Virginia to help assess radiative forcing by urban aerosols [81].

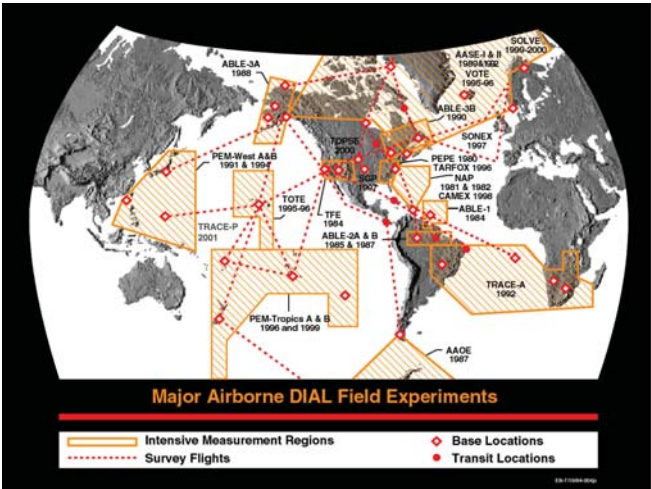


**Table 13.1.** Parameters of the NASA Langley airborne UV DIAL system (Courtesy of E.V. Browell, NASA LaRC)

|                                          |                                                                        |            |            |
|------------------------------------------|------------------------------------------------------------------------|------------|------------|
| Lasers:                                  | Continuum Model ND 6000 flashlamp-pumped Nd:YAG, Model 9030 dye lasers |            |            |
| Pulse repetition frequency/Hz            | 30                                                                     |            |            |
| Pulse length/ns                          | 8–12                                                                   |            |            |
| Pulse energy/mJ at: 1.06 $\mu\text{m}$   | 250–350                                                                |            |            |
| 600 nm                                   | 50–70                                                                  |            |            |
| 290/300 nm                               | 30                                                                     |            |            |
| Wavelength separation for ozone/nm       | 10                                                                     |            |            |
| Dimensions ( $l \times w \times h$ in m) | $5.699 \times 1.016 \times 1.092$                                      |            |            |
| Weight/kilograms                         | 1739                                                                   |            |            |
| Power requirements/kW                    | 30                                                                     |            |            |
| Receiver:                                | Wavelength Regions                                                     |            |            |
|                                          | 289–300 nm                                                             | 572–600 nm | 1064 nm    |
| Area of receiver/ $\text{m}^2$           | 0.086                                                                  | 0.086      | 0.086      |
| Receiver efficiency to detector/%        | 31                                                                     | 40         | 31         |
| Detector quantum efficiency/%            | 26 (PMT)                                                               | 8 (PMT)    | 40 (APD)   |
| Total receiver photon efficiency/%       | 6.5                                                                    | 3.2        | 12.4       |
| Receiver field of view (selectable)/mrad | $\leq 1.5$                                                             | $\leq 1.5$ | $\leq 1.5$ |

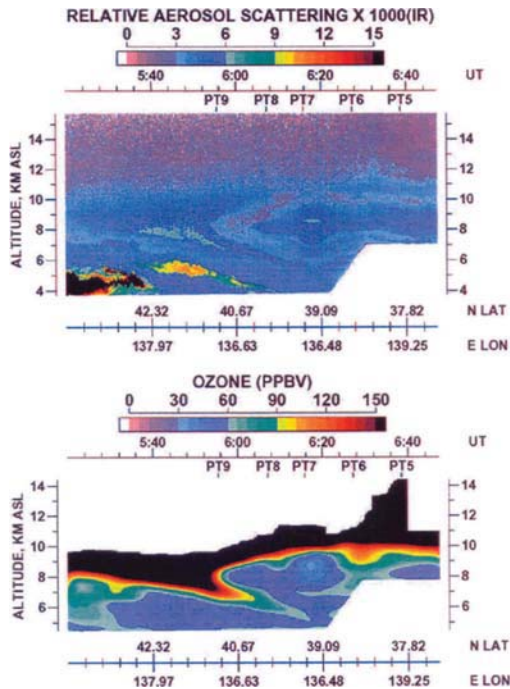


**Fig. 13.6.** An example of simultaneous O<sub>3</sub> and aerosol data taken with the LaRC airborne DIAL system. (Courtesy of E.V. Browell.)

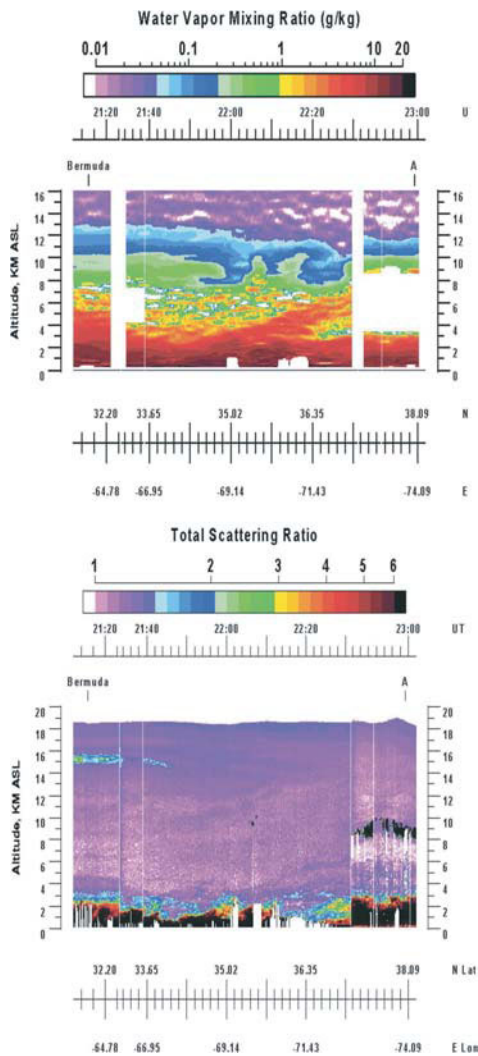


**Fig. 13.7.** An example of the simultaneous NASA LaRC LASE DIAL H<sub>2</sub>O and total scattering data taken aboard the NASA ER-2 aircraft. (Courtesy of E.V. Browell.)

## 13.5 The Use of Spaceborne Lidars

### 13.5.1 The LITE Experience

Quite separate of EOS, however, a proof-of-principle lidar experiment was approved by NASA for flight aboard Shuttle in the late 1980s. Owing to the Challenger mishap, the program was delayed, and the launch postponed until 1994. The launch from NASA's Kennedy Space Center

of the Lidar In-space Technology Experiment (LITE), which was the prime payload aboard the Space Shuttle Discovery flight STS-64, took place on September 9, 1994, at 6:23 p.m. EDT, after a 1 hour 53 minute delay due to unsatisfactory weather conditions. A crew of six astronauts was chosen for this 10-day mission. The orbit altitude of STS-64 was 259 km with an inclination of  $57^\circ$ . This orbit provided coverage over most of the Earth's surface allowing many different geophysical targets to be studied along with extensive opportunities for ground-truthing over many lidar sites. The late afternoon launch provided optimum viewing conditions for an extensive European validation campaign.

The scientific investigations planned for LITE are given by McCormick et al. [82] and McCormick [83]. Details of the LITE instrument were given in three papers and two posters at the 1992 Sixteenth International Laser Radar Conference (ILRC), Cambridge, MA, [84–88] and in two papers at the 1994 Seventeenth International Laser Radar Conference (ILRC) at Sendai, Japan [89, 90]. LITE employed a three-wavelength Nd:YAG laser transmitter and 1 m diameter telescope receiver with photomultipliers (PMTs) for the 355 nm and 532 nm channels, and an avalanche photodiode (APD) for the 1064 nm channel. A two-laser 10 Hz flashlamp pumped design was incorporated for redundancy. Laser energies at 1064, 532, and 355 nm were approximately 445, 557, and 162 mJ, respectively. The field of view of the receiver was determined by a selectable aperture stop: either a wide field of view at 3.5 mrad for nighttime measurements, a narrow aperture at 1.1 mrad for daytime measurements, or an annular field of view for multiple scattering measurements could be chosen. In addition, an opaque aperture could be inserted to protect the detectors when not in use. Narrowband interference filters were also moved into the optical path to reject the bright sunlit background during daytime portions of the orbits. The lidar return signals were amplified, digitized, stored on tape on board the Shuttle, and simultaneously telemetered to the ground for most of the mission using a high-speed data link.

An electronic bandwidth of 2 MHz limited the range resolution to about 35 m. The instrument was commanded from the ground over a low-rate telemetry link. In this manner, various instrument configuration modes were changed quickly during flight. Such things as photomultiplier high voltages, electronic amplifier gains, field of view, and optical filtering were routinely changed. A detailed mission plan and supporting correlative measurements plan was painstakingly developed for the 10-day mission. Nominally, LITE took data during ten 4-1/2 hour data taking sequences and, in addition, five 15-minute “snapshots” over

specific target sites. Toward the end of the mission, other data sequences were permitted. For example, data were taken during the night portions of five consecutive orbits (146–150) so that more spatial continuity could be obtained. Six aircraft carrying a number of up-looking and down-looking lidars performed validation measurements by flying along the LITE footprint. In addition, ground-based lidar and other measurements, e.g., balloon-borne dustsondes, were coordinated with LITE overflights. Photography took place from Shuttle during daylight portions of the orbits. A camera fixed and boresighted to LITE took pictures, as did the astronauts using two Hasselblad cameras and one camcorder to support LITE's lidar measurements. Prior to the flight, the astronauts were briefed on what targets of opportunity were of interest. Various cloud types, taken during previous shuttle flights, were used as examples.

Figure 13.8 shows the nearly completed LITE instrument in a clean room at NASA's Langley Research Center, where LITE was built. Atmospheric tests were conducted at LaRC before shipment (see Fig. 13.9) and again at NASA Kennedy Space Center (KSC) before Integration onto the Shuttle Discovery. Figure 13.10 shows LITE integrated into the shuttle Discovery in the Vehicle Assembly Building (VAB) at KSC. Figure 13.11 is a picture of the launch of LITE, and Fig. 13.12 shows LITE in orbit.

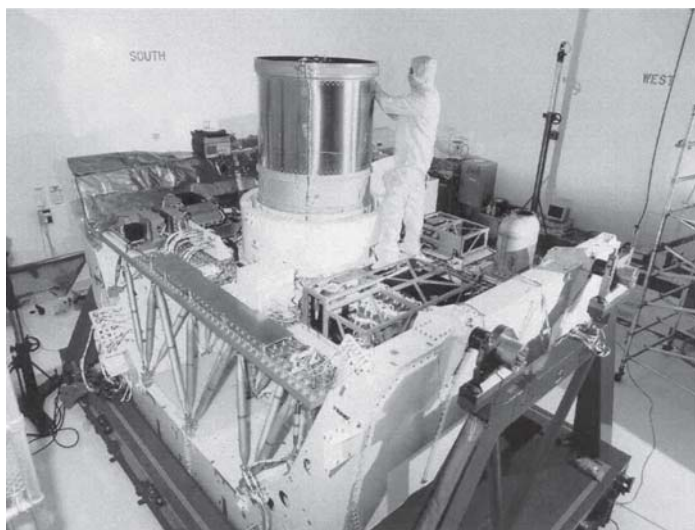


**Fig. 13.8.** NASA LaRC's LITE instrument being developed in a clean room at LaRC. The worker is shown next to the nearly 1-meter diameter receiving telescope.



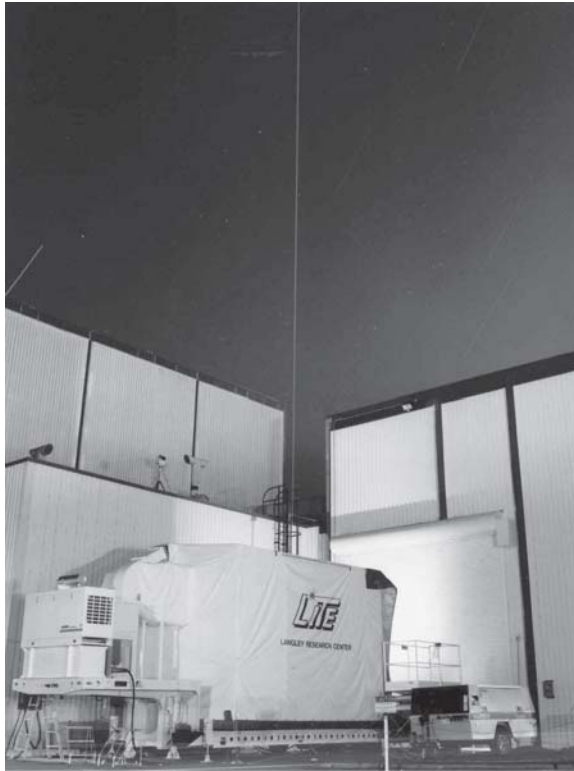


**Fig. 13.9.** LITE before shipment to NASA KSC for launch. Atmospheric tests being conducted at LaRC. A time exposure photograph shows the vertical green “pencil” beam from the Nd:YAG laser.

The LITE instrument was activated approximately 3 hours after launch and it met all expectations. Both lasers were utilized for a total of 1,929,600 shots. Although some degradation in laser output energy was observed as expected, both lasers continued to produce more than acceptable levels of energy throughout the mission. The only significant perturbation to pre-mission plans was caused by the inability to record high-rate data aboard the shuttle. As a result, adjustments to shuttle altitude were made to maximize Ka band coverage, so that high-rate data could be downlinked and recorded at NASA’s Johnson Space Center (JSC). Also, a science extension day was granted and replanned to acquire many nighttime half orbits, so that all science objectives could be met. Over 53 hours of 100-shot-averaged science data, and over 45 hours of high-rate science data were acquired during the 10-day LITE



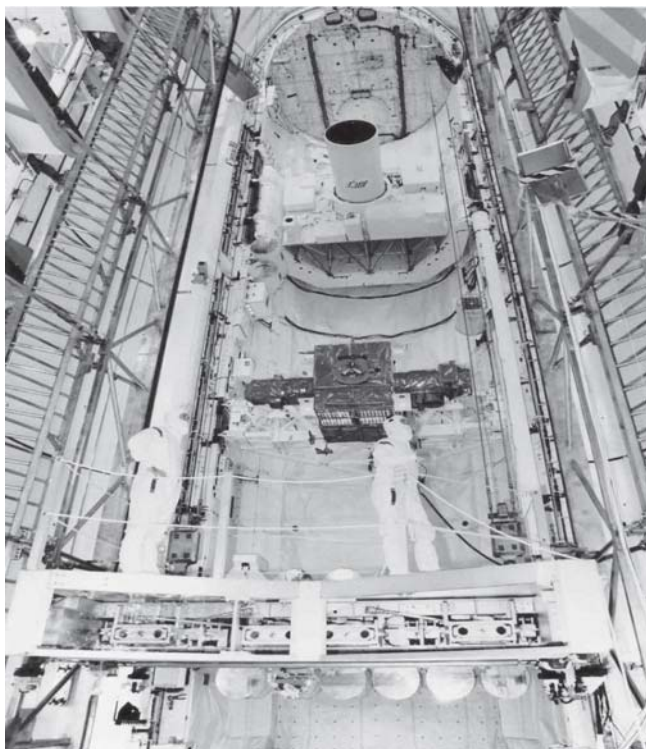


**Fig. 13.10.** LITE integrated into Shuttle at NASA's Kennedy Space Center.

flight. Additional background and noise data were also obtained to help with post-flight data analysis.

The raw data showed immediately the tremendous potential for a spaceborne lidar. Desert dust layers, biomass burning, pollution out-flow off continents, stratospheric volcanic aerosols, and many storm systems were observed. Complex cloud structures were observed over the intertropical convergence zone (ITCZ), with LITE penetrating the uppermost layer to four and five layers below. A great deal of stratus was observed over ocean areas. LITE showed clearly the effects of optically thick clouds on lidar penetration and measurement capability. LITE showed that space lidars could penetrate to altitudes of within 2 km of the surface 80% of the time, and reach the surface 60% of the time [91]. It appears that clouds with optical depths as high as 5–10 can be studied with lidars. Surface reflectance and multiscatter measurements were also conducted. A fortuitous event occurred when LITE passed directly over the eye of the Super Typhoon Melissa. Figure 13.13 shows that LITE



**Fig. 13.11.** The launch of the Shuttle Discovery with LITE aboard, September 9, 1994, at 1823 EDT. (Courtesy of NASA KSC.)

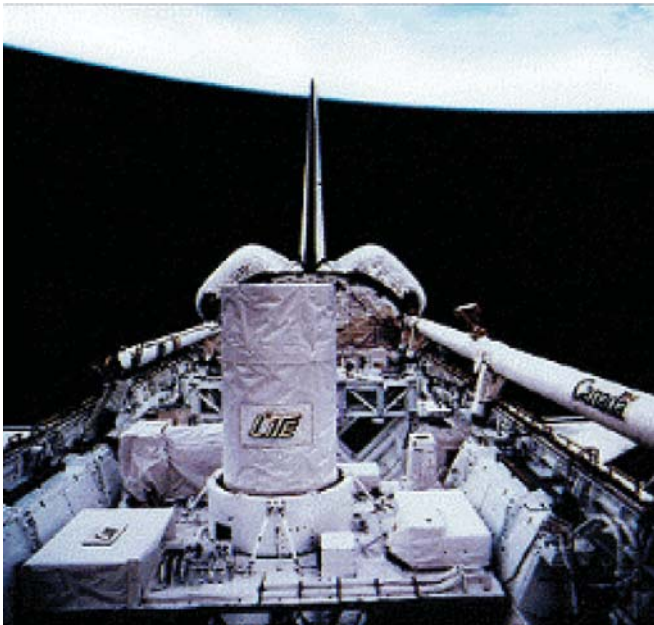


**Fig. 13.12.** LITE shown aboard Discovery in Earth orbit, September 1994.

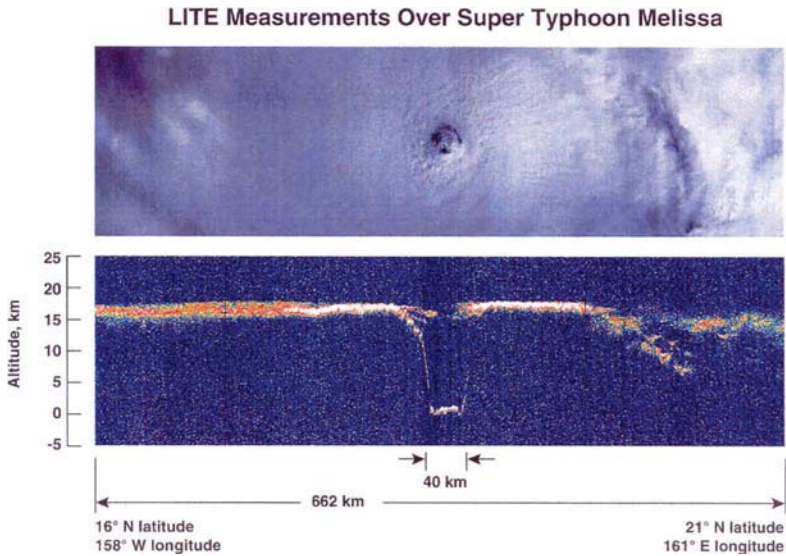


**Fig. 13.13.** A picture of the Super Typhoon Melissa taken by the LITE camera that is boresighted with the lidar (top panel). The bottom panel shows the attenuated 532-nm laser backscatter as LITE took data directly over the eye of the typhoon. These data were taken during daylight.

profiled the eye-wall cloud down to the surface [92]. Other data examples are shown in Figs. 13.14–13.17. As an example of studies associated with the LITE data, Fig. 13.18 uses 5-day back trajectories at about 1.5 km height to determine where the air masses of Fig. 13.17 originated. (Courtesy of C.R. Trepte, NASA LaRC).

A large correlative measurements program was coordinated to help in the validation of LITE. Observations were made at over 90 ground sites in 20 countries. For example, a NASA P-3B aircraft flew five correlative flights over the ground track of the Shuttle: (1) Wallops Island, VA, to Barbados, (2) Barbados to Ascension Island, (3) Ascension to Cape Town, S. Africa, (4) Cape Town to Ascension, and (5) Ascension to the Azores. On board the P-3 were two lidars, one zenith viewing to observe upper troposphere clouds and stratospheric aerosols (a LaRC lidar); and a second (a GSFC lidar) called LASAL, Large Aperture Scanning Airborne Lidar [93]. Figure 13.19 shows a time exposure photograph of the laser emissions from these systems. LASAL observed the 3-D structure of the planetary boundary layer (PBL) and low-level clouds. LASAL is a down-looking elastic backscatter lidar with a Nd:YAG laser transmitter and a 55 cm aperture telescope, in conjunction with a full aperture scan

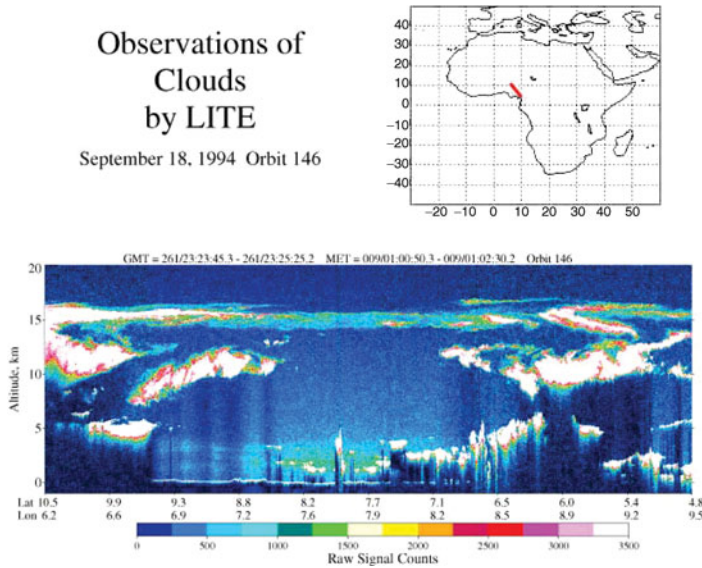


**Fig. 13.14.** An example of multiple cloud layers and cloud types in the Inter-Tropical Convergence Zone over Africa. LITE was able to penetrate a significant percentage of these clouds. Some were very optically thick preventing measurements to the surface. Multiple scattering effects allowed greater penetration but greatly complicated the measurement of the true cloud base. (Courtesy of K.A. Powell.)

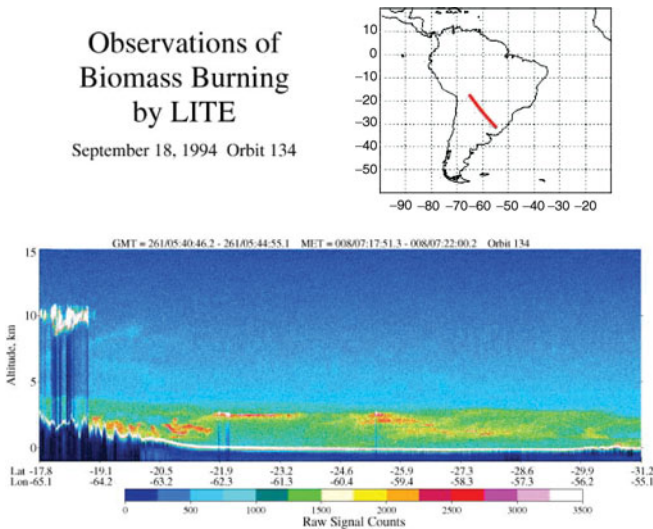


**Fig. 13.15.** An example of LITE observations of biomass smoke aloft. (Courtesy of K.A. Powell.)



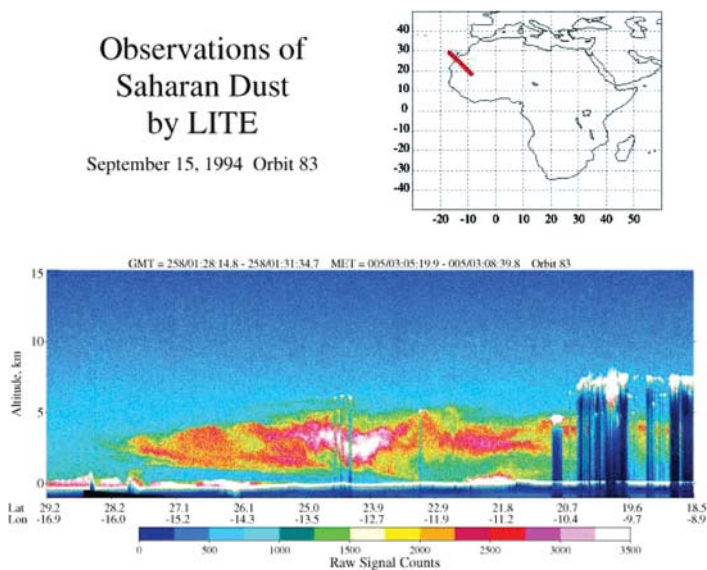


**Fig. 13.16.** This example of LITE data shows a plume of Saharan dust up to about 5 km altitude. (Courtesy of K.A. Powell.)

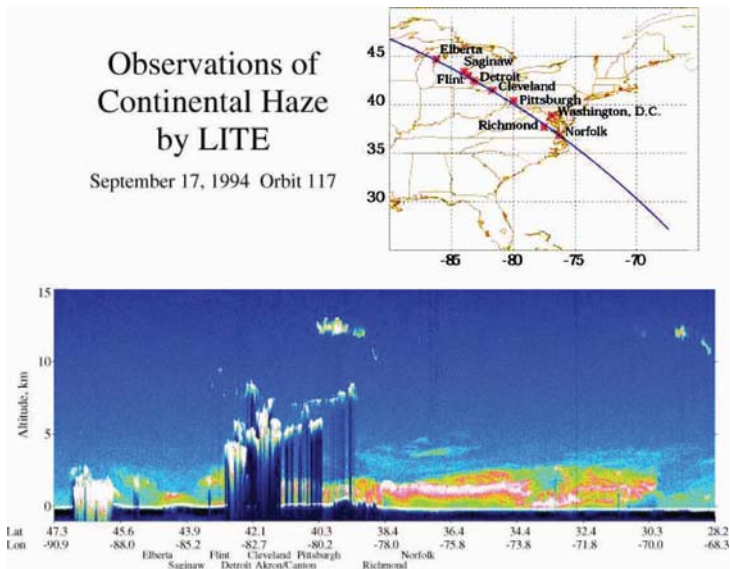


**Fig. 13.17.** A LITE observation of industrial/urban outflow over the eastern U.S. into western Atlantic Ocean. (Courtesy of K.A. Powell.)

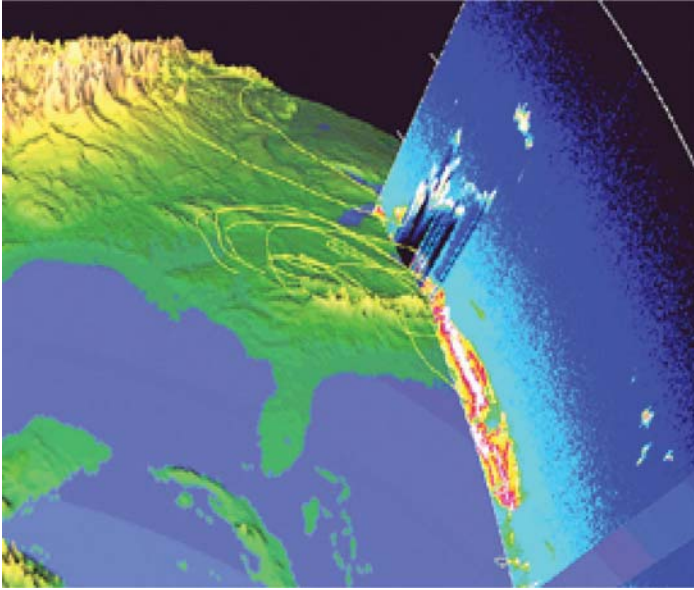


**Fig. 13.18.** LITE data showing a deep haze layer over the eastern U.S. and western Atlantic Ocean. Back trajectories show transport at the 850-mb level over the previous 5 days indicating that the cleaner air is from Canada and the higher aerosol concentrations have a source in the Ohio valley. The topography is greatly exaggerated in this figure, as is the vertical scale of the LITE data relative to the underlying map of the eastern U.S. (Courtesy of C.R. Trepte.)

mirror to measure aerosols and clouds in three dimensions. It is capable of cross track scanning  $\pm 45^\circ$  at scan rates up to  $90^\circ/\text{s}$ , and up to  $40^\circ$  fore of nadir along the flight track. LASAL provided a unique set of measurements to study the relationship of PBL structure and growth with ocean surface characteristics that can be related to surface fluxes of heat and moisture. The comparison of the Shuttle lidar data and the lidar data acquired on board the aircraft is remarkable (Fig. 13.20), with each showing nearly identical cloud layering, PBL structure, and lower tropospheric aerosol distributions [94]. Three other airborne lidar systems were used in the LITE validation program. A Canadian aircraft was flown off the coast of and over California [95]. One technique applied to LITE data analysis showed that airborne lidar measurements of size distribution and backscatter-to-extinction ratios from the literature could be used to estimate sulfate emission rates from industrial/urban regions [96]. Another example of a comparison between an airborne lidar and LITE tropospheric aerosol data is shown in Fig. 13.21. These data,



**Fig. 13.19.** NASA LaRC and GSFC lidars aboard the NASA P-3 for LITE validation. Downlooking beams represent a time-lapse photograph of the LASAL lidar fan beam system. The uplooking green line shows the LaRC 532 nm lidar output. (Courtesy of G.K. Schwemmer.)

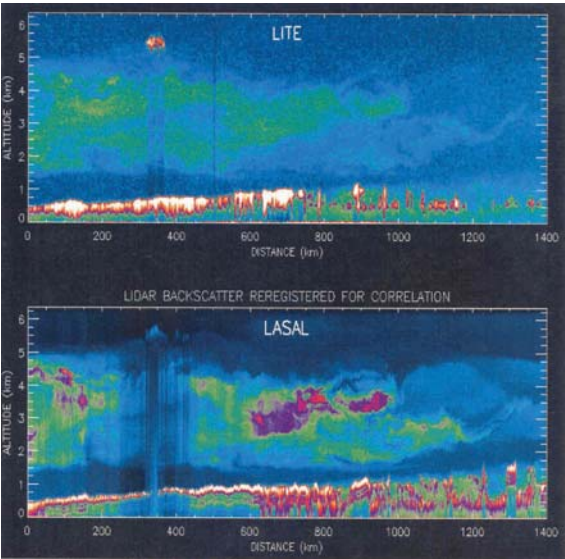


**Fig. 13.20.** A comparison of coincident LASAL and LITE data. (Courtesy of G.K. Schwemmer.)

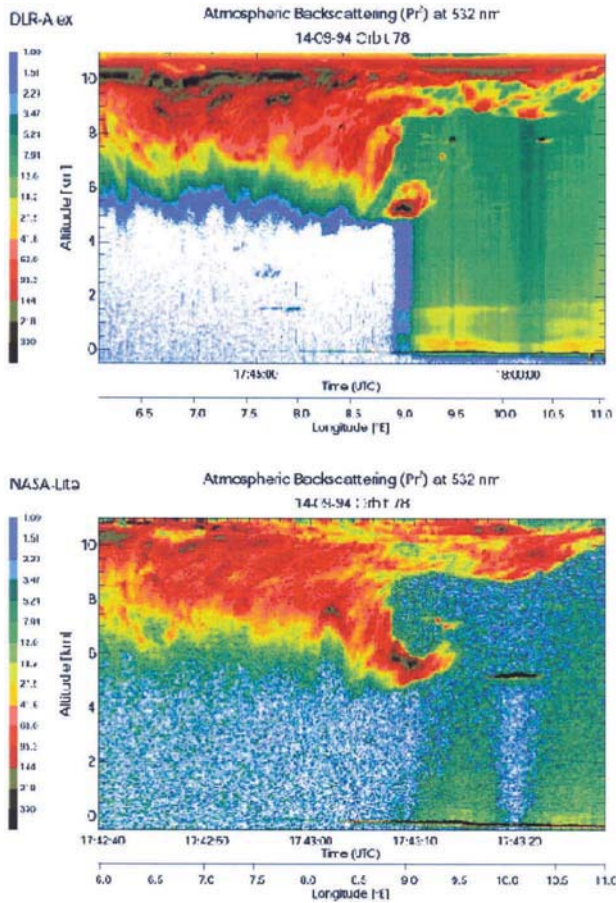


**Fig. 13.21.** Coincident airborne ALEX and LITE comparison data over Europe. (ALEX data courtesy of Ch. Werner.)

during orbits 78 and 79, compare the German DLR ALEX downlooking lidar [97] with LITE. The comparisons were made over Europe and the agreement is remarkable.

The STS-64 mission ended with landing at Edwards Air Force Base on September 20, 1994, at 5:13 p.m. EDT. The 11-day Shuttle flight of LITE did indeed usher in a new era of remote sensing the Earth's atmosphere from space. This flight showed the science community the exceedingly important data on clouds and aerosols that a spaceborne lidar can provide [98]. Many papers were written using the LITE data. Some of the validation and application papers include: Kent et al. [99] Menzies et al. [100]; Osborn et al. [101]; Strawbridge and Hoff [95]; Winker



and Poole [102]; Winker and Trepte [103]; Hoff and McCann [104]; Cuomo et al. [105]; Gu et al. [106]; and, Karyampudi et al. [107].

Previously, lidar active remote sensing techniques in the U.S. were not able to receive approval and funding for space flight in competition with passive sensors, which have been used since satellites first orbited Earth. Passive sensors, however, have great difficulty with vertically resolving and uniquely determining tropospheric species as well as geophysical parameters like temperature. It was obvious that the innate characteristics of lidars would provide a small footprint on the ground, i.e., high horizontal resolution, very high vertical resolution, a high sensitivity to aerosol measurements, and an excellent discrimination against noise because of laser spectral purity. Perhaps most importantly, these characteristics allow lidars to probe between clouds and penetrate through optically thin clouds and, therefore, profile the troposphere. Technology initially held the lidars back from successfully obtaining long duration Earth-orbiting flights in the 1970s through the 1990s. Long-lifetime, laser power efficiency, cooling and weight issues had to be solved if lidars were to fly aboard long duration Earth-orbiting spacecraft. In the late 1980s and 1990s diode-pumped and long-lived Nd:YAG lasers, and light-weight optics and structures, changed drastically the feasibility for lidar flights. Coupled with successes of LITE, MOLA (Mars Orbiting Laser Altimeter) [108] and others, lidars became competitive for spaceborne missions. Therefore, when new flight opportunities in the U.S. presented themselves, CALIPSO and GLAS were accepted for flight through the proposal process. GLAS was launched in January 2003, and CALIPSO is on schedule for a 2005 launch.

### 13.5.2 ALISSA

On April 23, 1996, nearly two years after the LITE flight, the French-Russian ALISSA lidar [109, 110] was installed on the PRIRODA module, and launched from Baikonour, in Kazakhstan. It was attached to the MIR Platform on April 26. ALISSA was conceived in 1986 as a simple monochromatic backscatter Mie lidar for cloud altimetry. Its scientific objective was to describe the vertical structure of clouds and in particular to provide the absolute altitude of cloud top. Such information was seen to be used as a complement to geostationary satellite imagery and the aim was to demonstrate the interest of such instrumentation for meteorological purposes.

ALISSA was conceived and built in France by the Service d'Aéronomie with the support of the French Space Agency, CNES,

and the participation of the Russian RKK Energia, which assumed the responsibility of the PRIRODA module. The Russian Institute of Applied Geophysics was responsible for the lasers. The instrument was installed in the PRIRODA module in front of one of the two windows and when used is pointed toward the nadir. Its main feature is the fact that it uses four Nd-YAG lasers with nominal energy of 10 mJ each with a repetition rate of 50 Hz. This feature proved to be very useful in the test period, as only one of the lasers was used at any time, and far below its nominal energy; the effective energy during the sequences reported during the tests was about 5 mJ at 532 nm.

The ALISSA instrument was used in a test mode during 18 sequences of 7 to 25 minutes duration from September 1996 to May 1997. Owing to difficulties with the MIR, measurements were interrupted until May 1999. Twenty-five more sequences were then acquired between May 7 and July 4, 1999—before the MIR was brought back to earth. The crew was able to change lasers from time to time in order for these data to be taken. The ALISSA lidar soundings have been compared with the corresponding IR images of the geostationary satellites METEOSAT, GOES, and GMS, with a temporal coincidence of 30 minutes for most of them. These comparisons are quite satisfactory at least on the main features of the cloud coverage, but one has to get better localization of the lidar soundings to go further. The observations of the lidar back-scattered signals showed the detection of multilayered clouds, high altitude cirrus, orographic waves, and the detection of the boundary layer. This demonstration, after the success of the preceding flight of LITE, showed that even a low power lidar that could be easily carried on an operational satellite could provide useful information for cloud description, including the description of the aerosol boundary layer.

### 13.5.3 GLAS

The first long-duration spaceborne lidar is the Geoscience Laser Altimeter System (GLAS) (see Fig. 13.22). GLAS is a facility instrument designed to measure ice-sheet topography and associated temporal changes, as well as cloud and atmospheric properties [111]. In addition, operation of GLAS over land and water will provide along-track topography. GLAS is carried on the Ice, Cloud and land Elevation Satellite (ICESat), which launched 13 January 2003 at 00:45 UTC from Vandenberg Air Force Base in California. GLAS incorporates a diode-pumped Nd:YAG laser to make surface topography measurements at 1064 nm. The measurement of aerosols and other atmospheric



**Fig. 13.22.** Shows the ICESat during fabrication in a clean room at Ball Aerospace and Technologies Corporation, Boulder, Colorado. ICESat was launched on a Boeing Corporation Delta II launch vehicle, January 2003.

characteristics is accomplished at the 532 nm wavelength. The return signal is collected using a 1-meter-diameter telescope. The laser transmits forty 5-ns pulses per second at the nadir producing 70-m-diameter spots at the surface, separated by 175 meters. Figure 13.23 is an artist's rendition of the ICESat on orbit.

The GLAS atmospheric measurements are obtained from the ICESat 600-km polar orbiting platform both day and night. The 532-nm photon counting channel employs an etalon filter that is actively tuned to the laser frequency, providing a bandpass filter of about 30 picometers full width. This, together with a very narrow ( $160\ \mu\text{rad}$ ) receiver field of view (FOV), should allow high-quality daytime measurements even over bright background scenes. There are eight separate photon counting detectors for the 532-nm channel, which will significantly increase the available dynamic range, while providing some degree of redundancy in



**Fig. 13.23.** An artist's rendition of the ICESat spacecraft with the GLAS instrument onboard. The 1064 nm and 532 nm laser pulses are shown probing the Earth's atmosphere and polar ice thickness changes. (Courtesy of S.P. Palm.)

the case of detector failure. The 1064-nm channel uses an avalanche photodiode (APD) detector with a much wider (1.0-nm) bandpass filter and FOV ( $475\ \mu\text{rad}$ ). The sensitivity of the 1064-nm channel is limited by the inherent detector noise. It should, however, provide sufficient signal-to-noise ratio to profile most clouds and some of the denser aerosol layers. It will also be used to supplement the 532-nm channel when, and if, it becomes saturated. The 1064-nm data will not be used to retrieve atmospheric parameters since the signal-to-noise ratio of the 532-nm channel will be much better. The only exception to this would be in the case of problems, or complete failure, of the 532-nm channel, at which point the 1064-nm channel data would be used for cloud height retrieval only. Table 13.2 lists the major GLAS system parameters that ultimately affect system performance and data quality.

GLAS carries three identical and redundant 40 Hz, solid-state Nd:YAG lasers on board, each with an expected lifetime of about 2.5 billion shots, or approximately two years of continuous operation. If a laser malfunctions, or simply comes to the end of its normal lifetime, switching to one of the other lasers is straightforward. The backscattered light from atmospheric clouds, aerosols and molecules is digitized at 1.953 MHz, yielding a vertical resolution of 76.8 m. The horizontal resolution is a function of height. For the lowest 10 km, each backscattered

**Table 13.2.** System parameters for the spaceborne GLAS lidar (Courtesy of S.P. Palm.)

| Parameter                    | 532-nm Channel          | 1064-nm Channel        |
|------------------------------|-------------------------|------------------------|
| Orbit altitude               | 600 km                  | 600 km                 |
| Laser energy                 | 36 mJ                   | 73 mJ                  |
| Laser divergence             | 110 $\mu$ rad           | 110 $\mu$ rad          |
| Laser repetition rate        | 40 Hz                   | 40 Hz                  |
| Effective telescope diameter | 0.95 m                  | 0.95 m                 |
| Receiver field of view       | 160 $\mu$ rad           | 475 $\mu$ rad          |
| Detector quantum efficiency  | 70%                     | 35%                    |
| Detector dark current        | $3.0 \times 10^{-16}$ A | $50 \times 10^{-12}$ A |
| RMS detector noise           | 0.0                     | $20 \times 10^{-12}$ A |
| Electrical bandwidth         | $1.953 \times 10^6$ Hz  | $1.953 \times 10^6$ Hz |
| Optical filter bandwidth     | 0.030 nm                | 1.00 nm                |
| Total optical transmission   | 30%                     | 55%                    |

laser pulse will be stored resulting in a horizontal resolution of 175 m. Between 10 and 20 km altitude, eight shots will be summed, producing a horizontal resolution of 1.4 km. For the upper half of the profile (20–40 km), which is entirely within the stratosphere, 40 shots will be summed, providing a horizontal resolution of about 7 km. This approach was adopted for a number of reasons. First, the atmospheric processes of interest have more variability and smaller scales in the lower troposphere (particularly the boundary layer) than in the mid-and-upper troposphere. Second, the amount of molecular and aerosol scattering in the upper troposphere and stratosphere is so small that summing multiple shots is required to obtain a non-zero result. Lastly, this approach will help to reduce the amount of data that has to be stored on board the spacecraft and transmitted to the ground.

13.5.4 CALIPSO

Recent assessments by the international science community conclude that better global observations of clouds and aerosols are required in order to reduce uncertainties in predictions of future climate change. The CALIPSO mission (previously known as PICASSO) is a satellite experiment designed to provide new observational capabilities that will fill measurement gaps in the current NASA Earth Observing System. It will provide vertical profiles of clouds and aerosols and their properties, which will help address current uncertainties in aerosol and cloud effects on earth radiation budget. It uses a three-channel lidar, three-channel infrared radiometer, and a wide-field camera on a French Proteus

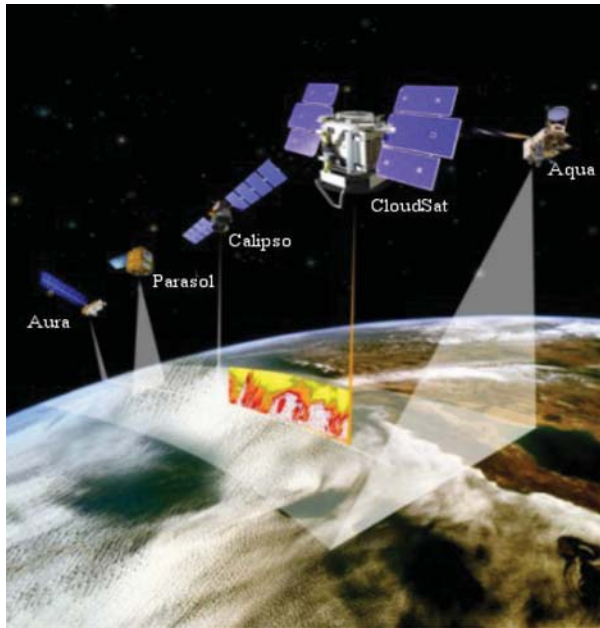


**Fig. 13.24.** Five satellites that will fly in formation during the CALIPSO mission are shown. This grouping is known as the EOS Aqua constellation or “A-train.” The “A-train” name comes from the old jazz tune, “Take the A-Train” composed by Billy Strayhorn and made popular by Duke Ellington’s band. It is an Afternoon constellation and has Aqua in the lead with Aura in the rear. (Courtesy of NASA GSFC.)

spacecraft flying in formation with Aqua (previously known as EOS-PM), CloudSat, AURA, and Parosol. Figures 13.24 and 13.25 show this constellation of satellites that together will allow numerous synergies to be realized [112]. The launch is scheduled for 2005 with a three-year minimum duration [113, 114]. CALIPSO is a cooperative effort led by NASA’s Langley Research Center and includes CNES, Hampton University, Ball Aerospace and Technologies Corporation, and the French Institut Pierre Simon Laplace. The five satellites will produce an unprecedented and comprehensive suite of nearly coincident measurements of atmospheric state, aerosol and cloud optical properties, and radiative fluxes. These data will allow fundamental advances in our understanding of future climate change.

The three nadir-viewing instruments that will fly on CALIPSO are: the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), the Imaging Infrared Radiometer (IIR), and the Wide Field Camera (WFC). These instruments are designed to operate autonomously and

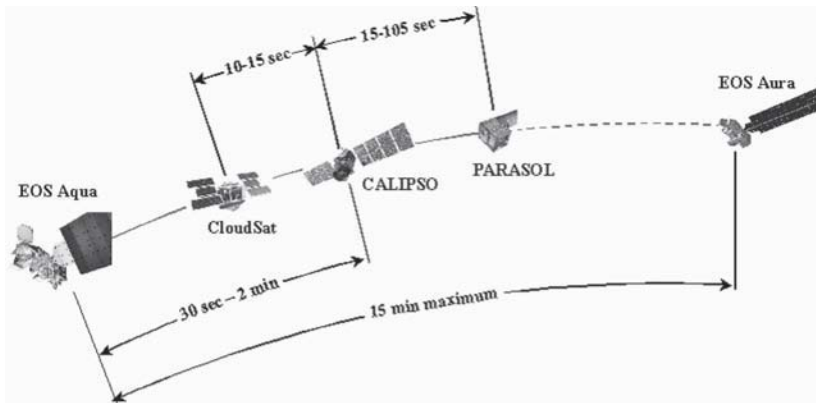


**Fig. 13.25.** The five satellites of the EOS Aqua constellation shown in formation with time differences between the satellites and, therefore, their measurements. This arrangement is being adjusted as the various measurements are being studied and will change. An issue, for example, is sunglint and MODIS measurements, which has caused CALIPSO and CloudSat to make orbit adjustments. (Courtesy of L.R. Poole.)

continuously, except for the WFC that acquires science data only under daylight conditions. Science data are downlinked using an X-band transmitter system that is part of the payload. The physical layout of the payload is shown in Fig. 13.26, with key instrument characteristics listed in Table 13.3.

CALIOP utilizes a diode-pumped Nd:YAG laser and frequency doubler which produces linearly polarized pulses of light at 1064 nm and 532 nm. The atmospheric return is collected by a 1-meter telescope that directs the photons to a three-channel receiver measuring the backscattered intensity at 1064 nm, and the two orthogonal polarization components at 532 nm (parallel and perpendicular to the polarization plane of the transmitted beam). The receiver subsystem consists of the telescope, relay optics, detectors, preamplifiers, and line drivers mounted on a stable optical bench. Signal processing and control electronics are contained in boxes mounted on the payload housing. The receiver telescope is an all-beryllium 1-meter-diameter design similar to the telescope built for GLAS. The telescope primary mirror, secondary mirror, metering structure, and inner baffle are all made of beryllium. A carbon composite light shade is added to prevent direct solar illumination of the mirrors. A fixed field stop is located at the telescope focus. A mechanism located in the collimated portion of the beam contains a shutter and a depolarizer used in calibrating the 532 nm perpendicular channel. A narrowband etalon is used in the 532 nm channel to reduce the solar



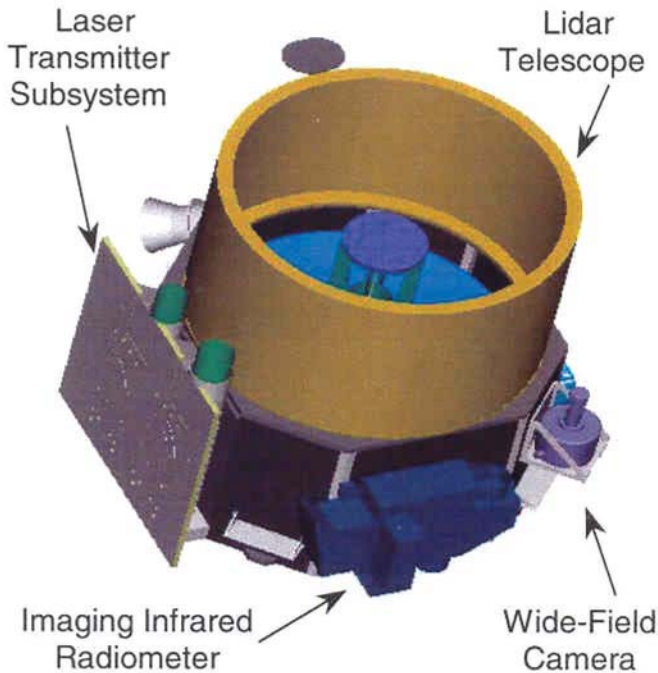


**Fig. 13.26.** An artist's rendition of the placement of instruments aboard the CALIPSO spacecraft. (Courtesy of D.M. Winker.)

background illumination, while a dielectric interference filter provides sufficient solar rejection for the 1064 nm channel. Dual digitizers on each channel provide an effective 22-bit dynamic range needed to measure both cloud and molecular backscatter signals. An active beam steering system ensures alignment between the transmitter and the receiver. The laser transmitter subsystem consists of two redundant lasers, each with a beam expander, and the beam steering system. The Nd:YAG lasers are Q-switched and frequency-doubled to produce simultaneous pulses at 1064 nm and 532 nm. Each laser produces 110 mJ of energy at each of the two wavelengths at a pulse repetition rate of 20.2 Hz. Only one laser is operated at a time. Beam expanders reduce the angular divergence of the laser beam to produce a beam diameter of 92 meters at the Earth's surface. The lasers are passively cooled using a dedicated radiator panel, avoiding the use of pumps and coolant loops. Each laser is housed in its own sealed canister filled with dry air at standard atmospheric pressure.

The fundamental sampling resolution of the lidar is 30 m vertical and 333 m horizontal, determined by the receiver electrical bandwidth and the laser pulse repetition rate. Backscatter data will be acquired from the



**Table 13.3.** System parameters for the CALIPSO space-craft mission to be launched in 2005. The characteristics for the lidar (CALIOP), wide field camera (WFC), and the Imaging Infrared Radiometer (IIR) are shown (Courtesy of D.M. Winker.)

| Characteristic          | Value            |         |
|-------------------------|------------------|---------|
| CALIOP                  |                  |         |
| Wavelengths (μm)        | 0.532 and 1.064  |         |
| Pulse energy (mJ)       | 110 at each λ    |         |
| Polarization            | 0.532 ∥ and ⊥    |         |
| Resolution at alt. (km) | ΔZ (m)           | ΔH (km) |
| 0–8                     | 30               | 0.33    |
| 8–20                    | 60               | 1       |
| 20–30                   | 80               | 1.67    |
| 30–40                   | 300              | 5       |
| WFC                     |                  |         |
| Wavelength (μm)         | 0.645            |         |
| Δλ (μm)                 | 0.05             |         |
| IFOV (km)               | 0.125            |         |
| Swath (km)              | 61               |         |
| IIR                     |                  |         |
| Wavelengths (μm)        | 8.65, 10.6, 12.0 |         |
| Δλ (μm)                 | 0.6 to 1.0       |         |
| IFOV (km)               | 1                |         |
| Swath (km)              | 64               |         |

surface to 40 km with 30 m vertical resolution. Low-altitude data will be down linked at full resolution, but to reduce the required telemetry bandwidth, data from higher altitudes will be vertically and horizontally averaged on board. The lidar profiles are averaged to a resolution of 60 m vertical and 1 km horizontal from 8 to 20 km, 80 m vertical and 1.67 km horizontal from 20 to 30 km, and 300 m vertical and 5 km horizontal from 30 to 40 km. The lidar is calibrated by normalizing the return signal at altitudes between 30 km and 35 km. A depolarizer can be inserted into the 532-nm beampath to calibrate the perpendicular channel relative to the parallel channel. The 1064-nm channel is calibrated relative to the 532-nm total backscatter signal using cirrus clouds as targets.

A comprehensive validation effort is planned for CALIPSO. It will include global campaigns as well as routine measurement comparisons with existing networks of lidars, sun photometers, and other measurements. The validation plan also includes international quid pro quo measurements in countries throughout the world [115].

Not only did LITE demonstrate the potential of space lidar for observation of clouds and aerosols, but it also has been exceedingly important for performing simulations of how CALIPSO will work in orbit. Figure 13.27 shows simulated CALIPSO 532-nm raw data derived from LITE observations. It shows in the top left panel a LITE attenuated backscatter measurement at 532 nm taken on September 18, 1994. Smoke is evident below 5 km (golden color) created by biomass burning, and a number of cloud layers (white color) are also evident. The black vertical lines below the lowest clouds indicate that the backscatter was totally attenuated by the cloud. The data were taken over southwestern Africa during orbit 146. The clouds vary from subvisible cirrus near the tropopause at 17 km, to optically “thin” cirrus between the altitudes of approximately 10 km to 15 km, to highly attenuating clouds between about 5 km and 10 km, probably made of supercooled water droplets. These data provide a good test for CALIPSO simulations. The lower left panel depicts the 532-nm predicted capability of CALIPSO from its orbit to make the same measurement as LITE. As can be seen, CALIPSO will faithfully reproduce these data with its 532-nm channel. The upper right panel is for the 1064-nm channel and the lower right panel shows the

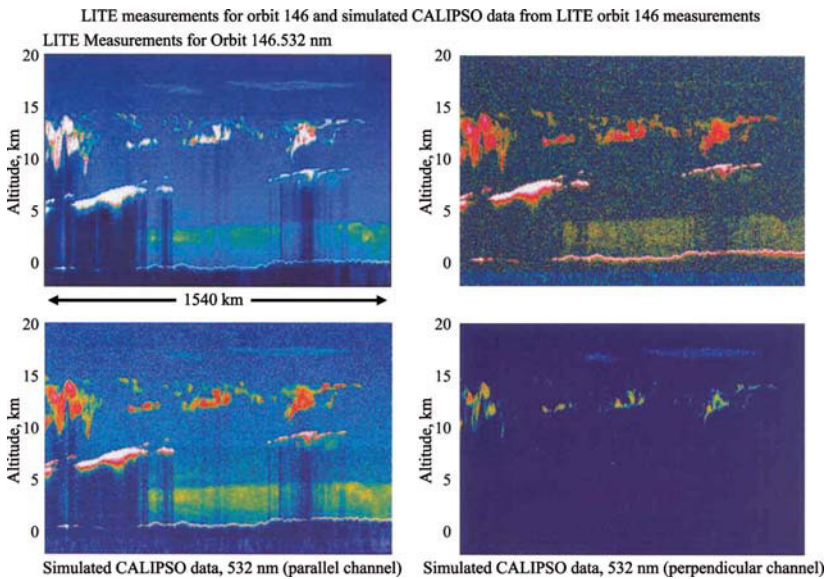


**Fig. 13.27.** CALIPSO simulations using LITE data as input for 532 nm parallel and perpendicular channels and for the 1064 nm channel. (Courtesy of K.A. Powell.)

532-nm perpendicular-polarization backscatter. The 1064-nm data and the 532-nm perpendicular-polarization data will help determine the type of aerosol present. For example, the depolarization data show that most of the 10–15 km cirrus clouds and much of the higher subvisible cirrus clouds are made of ice particles. CALIPSO will provide these capabilities over bright land surfaces, at night, and under conditions almost impossible for passive sensors to determine. Combining the CALIPSO capability with MODIS and CERES data will allow observationally-derived global estimates of aerosol forcing to be determined.

The satellites of the Aqua constellation will fly in a sun-synchronous, 705-km circular orbit with nominal ascending node equatorial crossing times ranging from 13:30 to 13:45 local (EOS Aqua). The orbits of CALIPSO, CloudSat, and PARASOL will be maintained to provide measurements that are nearly coincidental in space and time with those from Aqua, which was launched in May 2002. See Poole et al. [114] for more discussion on the measurement synergies that will be possible by these five satellite experiments flying in formation.

## 13.6 The Future

In the first decade of the twenty-first century, at least two long-duration lidars, GLAS and CALIPSO, will be circling the Earth, both with the goals to measure aerosols and clouds and use those observationally-derived data to improve climate modeling; for example, through a better estimate of climate forcing by aerosols. The addition of data from the Aura constellation, in the case of CALIPSO, will both be a challenge and an attribute. The challenge is to incorporate these data into a more complete and understandable data set, and to use the data for various modeling studies and for a more complete understanding of climate forcing. The synergy is that it offers a much more complete description of direct and indirect forcing, for example. The constellation, flying in formation, providing data unattainable by a single remote sensor, provides a paradigm for future satellite missions.

These first spaceborne long-duration missions utilize elastic backscatter only. Many studies, however, have taken place that show the feasibility of utilizing the DIAL technique to make measurements of ozone and water vapor. For example, Uchino et al. [116] showed that ozone could be measured in the 30–47 km altitude range with a vertical resolution of 1 km and a horizontal resolution of 800 km. If the vertical

resolution is relaxed to 3 km at 800 km horizontal resolution, the ozone uncertainties were shown to be reduced to about 3%. Mauldin et al. [117] gave a conceptual study for the EOS LASA (Lidar Atmospheric Sounder and Altimeter) facility that combined a backscatter aerosol and cloud experiment with a DIAL water vapor capability. It showed that water vapor could be measured from the surface to about 8 km with 5% accuracy. Spaceborne DIAL systems flying in this first decade are technologically feasible. However, NASA, NASDA and ESA chose an aircraft mission instead. Nevertheless, an ESA phase A spacecraft study is in progress which includes two lidar proposals out of a total of four potential missions. One is an H<sub>2</sub>O DIAL lidar and the other combines a lidar and radar. The flights of the two missions chosen for study are scheduled for 2008 and 2010. So it is possible that a DIAL system could be in orbit by the last year(s) of the first decade, as well as another CALIPSO-CloudSat type of mission.

Furthermore, as lasers provide more usable wavelengths for increased profiling capability with eye-safe operation, increase their repetition rates and lifetime, and become more energy-efficient, other applications will become possible from space. In addition to laser improvements, large deployable telescopes will enhance the capability of existing lidars and allow new applications. Improvements to ICESat and the next generation aerosol lidars are also being studied, and laser altimeters that can provide centimeter height resolutions are all possible by the end of the first decade.

Although outside the theme of this chapter, ESA has funded a wind lidar facility instrument, called the Atmospheric Laser Doppler Lidar (ALADIN) to fly aboard the AEOLUS spacecraft. It is being developed and scheduled for launch in 2007. ALADIN utilizes the Doppler shift in molecular backscatter associated with the wind profile. It measures in two wavelength bands on either side of the output frequency from the tripled wavelength of a Nd:YAG laser emitting at 355 nm [118–120].

The above improvements will enable future applications to be implemented in the subsequent decades like studies of the carbon cycle, circulation and forecasting through global tropospheric wind measurements, DIAL for constituent measurements and elastic backscatter for aerosol and cloud measurements. The implementation of these lidars in space will greatly enhance our understanding of atmospheric chemistry and climate. The future is indeed bright for spaceborne lidars, which are now taking their place alongside passive sensors, and fulfilling a myriad of measurement needs for the study of our Earth system.

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## DIAL Revisited: BELINDA and White-Light Femtosecond Lidar

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### 14.1 Introduction

Differential-absorption lidar or DIAL constitutes the first successful attempt to quantitatively measure concentration distributions of atmospheric trace gases in four dimensions. From a comparison of the capabilities of modern DIAL systems as those presented in Chapters 7 and 8 of this book with the pioneering work in the field (cf., e.g., [1]), the enormous progress the technique has made in the past thirty years becomes evident. Nevertheless there remains room for improvement.

One fact often neglected or ignored is the different degree of absorption of different parts of the laser spectrum in different parts of the absorption line. This effect is particularly important when the widths of the laser line and the absorption line of the gas of interest are comparable. It can lead to errors up to  $-50/ + 100\%$  in the case of near-IR water vapor DIAL and more than  $\pm 10$  K for temperature and must be considered in the data evaluation procedure.

Another point to consider is the technical problem to provide identical spatial structure of the output pulses and to precisely adjust the axes of two laser beams with one another and with the optical axis of the receiver (for two-laser DIAL systems), or to ensure identical beam directions

and perfect timing for the two output wavelengths (for switched-laser DIALs).

Both problems are eliminated or greatly reduced with a new approach best described by the expression “broadband-emission lidar with narrow-band determination of absorption,” or BELINDA. The BELINDA principle consists in short in the emission of laser pulses of spectral width about twice the width of the absorption line, and the use of two pairs of filters in the receiver, one transmitting close to line center and one in the wings of the lidar return signal. The filters close to line center provide what would be the on-resonance signal, the ones in the wings the off-resonance signal of a classical DIAL system. The first part of this chapter is devoted to the description of BELINDA, or “DIAL the other way around,” as it is sometimes called.

In the longer run, however, there are other improvements that appear highly desirable for a more comprehensive use of lidar in atmospheric research and pollution assessment. One is the extension to a larger number of atmospheric trace gases and the simultaneous measurement of the targeted pollutants. Conventional two-wavelength DIAL is restricted to the detection of a single pollutant at a time, leading to unacceptable delays in the measurement of a larger palette of pollutants. This is a major drawback in case of complex mixtures such as volatile organic compounds (VOCs) and photo-chemically active substances as occur, e.g., in the  $\text{O}_3\text{-NO}_x\text{-VOC}$  photocycle. Another is the detection of unexpected, unpredictable or unknown pollutants, released, for example, from hazardous exhausts, fires, unknown emitters, or in accidents. A third is the determination of chemical composition of not just the gaseous, but also the particulate constituents of the atmosphere. A fourth is the reduction of cross-sensitivities, or spectral interference, and increase in sensitivity and accuracy particularly for the less-abundant gases for which weak transitions of more abundant ones can overlap with the absorption features under investigation. A fifth, finally, is the search for and exploitation of a process that scatters predominantly backward, i.e., does not scatter all but a minute fraction of the laser radiation into directions where it is lost for the analysis, thus causing the  $1/(\text{distance})^2$  law with the ensuing rapid decrease of sensitivity, accuracy and depth resolution with distance.

In 1998 a group of lidar scientists carried out an experiment that gave hope that the above-mentioned auspices might be more than wishful thinking and that a solution could be at hand if it were possible to exploit

atmospheric white-light plasma channels [2, 3]. They shone intense ultrashort pulses from a high-power femtosecond laser (220 mJ, 100 fs) vertically into the sky and observed for the first time the phenomenon of white-light generation in an extended plasma channel in the atmosphere. Time-resolved measurements of the backscattered white light using a lidar setup showed backscatter signals from which atmospheric properties at altitudes as high as 12 km can be derived. This experiment opens a new field of lidar investigations: the white light femtosecond lidar technique. It is based on the nonlinear propagation of ultrashort and ultra-intense laser pulses in the atmosphere. Its important features are Kerr focusing of the beam followed by plasma defocusing; this leads to filamentation and to the generation of a broad spectral distribution (“white light”), with the resulting induced radiation emitted indeed primarily in the backward direction. The processes involved and some of the experiments carried out in connection with white-light femtosecond lidar will be the subject of the second part of this chapter.

## 14.2 BELINDA—*Broadband Emission Lidar with Narrowband Determination of Absorption*

### 14.2.1 Scattering Processes

The lidar return signal is due to scattering by air molecules and by cloud and aerosol particles. Whereas particle scattering can be treated as purely elastic, i.e., the backscattered spectrum is quasi-identical to the transmitted spectrum, the part of the backscattered spectrum that is due to molecular scattering is broadened. Following She [4] for the nomenclature of the different parts of the backscattered spectrum, only the central Cabannes line will be discussed here in some detail.

#### Doppler Broadening

The spectral width of the Cabannes line is determined by the motion of the scatterers. A single scattering event causes a spectral shift of the backscattered photon by an amount

$$\Delta\nu = 2 \cdot \nu_0 \cdot \frac{v_{\text{LOS}}}{c}, \quad (14.1)$$

where  $\nu_0$  is the wavenumber of the incident light,  $v_{\text{LOS}}$  the velocity of the scatterer along the line of sight, and  $c$  is the speed of light.

The velocities of an ensemble of scatterers at temperature  $T$  obey a Maxwell velocity distribution, leading to a backscattered spectrum of Gaussian type with half width (HWHM)

$$w_D = 2 \frac{v_0}{c} \sqrt{2 \cdot k_B \cdot T \cdot \ln 2 / M}, \quad (14.2)$$

$k_B$  being the Boltzmann constant and  $M$  the mass of the scatterers (mean mass of air molecules).

### Brillouin Scattering

The Maxwell velocity distribution holds only for scatterers in thermodynamic equilibrium. This condition is not met in the atmosphere because of pressure fluctuations induced for example by acoustic waves. The fluctuations cause the Cabannes line to split into a triplet [4–6]. Instead of considering in detail the exact shape of the Brillouin-modified return signal, a more pragmatic approach is usually taken in DIAL experiments. It consists in the assumption of a Gaussian distribution of the backscattered radiation with a modified width

$$w_D^{\text{cor}} = w_D \cdot \text{cor}, \quad (14.3)$$

$\text{cor}$  being an empirical correction factor that varies between 1.2 at ground level and 1.1 at 10 km height [7, 8]. The Rayleigh–Brillouin corrected spectrum of the backscattered radiation is henceforth denoted by  $\text{rbs}(v)$ .

## 14.2.2 Lidar Equations

### The Lidar Equation Including Broadening Effects

Until now the single-scattering elastic lidar equation had been written as

$$P(v, z, \beta) = P_0 \frac{c \Delta t}{2} \frac{A \eta O(z)}{z^2} \cdot \beta(z) \tau_\kappa^2(v, z) \tau_\alpha^2(z) \quad (14.4)$$

where  $P$  is the power received from range  $z$ ,  $P_0 c \Delta t$  is the laser pulse energy,  $A$ ,  $\eta$ ,  $O(z)$  are the receiver optics area, efficiency, and overlap integral,  $\beta$  is the backscatter coefficient, and  $\tau_\alpha = \exp \left[ - \int_0^z \alpha(v, z') dz' \right]$  and  $\tau_\kappa = \exp \left[ - \int_0^z \kappa(v, z') dz' \right]$  are the single-path extinction by the normal atmosphere and by the gas of interest, respectively. Equation (14.4)

neglects the broadening effects outlined in the previous section. To take care of those, Eq. (14.4) is modified to yield

$$P(\nu, z, \beta) = C(z, \beta) \cdot \tau_\kappa(\nu, z) \cdot B(\nu, z, \beta). \quad (14.5)$$

The quantity  $C$  summarizes all those factors on the right-hand side of Eq. (14.4) that do not depend on wavenumber and will cancel out later. The new factor

$$\begin{aligned} B(\nu, z, \beta) &= \frac{\beta_{\text{aer}}}{\beta} L(\nu, z) + \frac{\beta_{\text{mol}}}{\beta} (L(z) * rbs)(\nu) \\ &= [1 - x(z)] L(\nu, z) + x(z) (L(z) * rbs)(\nu) \end{aligned} \quad (14.6)$$

describes the changes due to the scattering process. It contains, in the (unbroadened) aerosol contribution, the extinction-weighted laser emission spectrum  $l(\nu)$ , i.e.,

$$L(\nu, z) = l(\nu) \tau_\kappa(\nu, z), \quad (14.7)$$

and in the broadened molecular contribution its convolution with the Rayleigh–Brillouin frequency distribution  $rbs$ ,

$$(L(z) * rbs)(\nu) = \int_{-\infty}^{\infty} L(\nu', z) \cdot rbs(\nu - \nu') d\nu'. \quad (14.8)$$

The reciprocal backscatter ratio  $x$  is given by

$$x = \frac{\beta_{\text{mol}}(z)}{\beta(z)} = \frac{\beta_{\text{mol}}(z)}{\beta_{\text{mol}}(z) + \beta_{\text{aer}}(z)}. \quad (14.9)$$

The unknown extinction coefficient  $\kappa$  appears now in both the second and third term of the right-hand side of Eq. (14.5).

### DIAL Inversion of the Lidar Equation with Broadening Effects

The absorption coefficient  $\kappa = N \cdot \sigma$  contains the information on either the molecule number density  $N$  which can be used for the determination of the concentration of a gas such as water vapor, or the absorption cross section  $\sigma$  from which the temperature can be derived [8, 9]. To obtain  $\kappa$  from the measured signals the quantity

$$Q(\bar{z}) = \ln \frac{P(\nu_1, z_1, \beta(z_1))}{P(\nu_2, z_1, \beta(z_1))} - \ln \frac{P(\nu_1, z_2, \beta(z_2))}{P(\nu_2, z_2, \beta(z_2))} \quad (14.10)$$

must be calculated for each height  $\bar{z} = (z_1 + z_2)/2$  along the lidar beam. To enhance readability  $\bar{z}$  will henceforth be  $z$  again. The on-resonance and off-resonance wavenumber of the absorption line are denoted by  $\nu_1$  and  $\nu_2$ , respectively. Insertion of Eq. (14.5) into Eq. (14.10) and some rearrangement yields

$$Q(z) = 2 \cdot \Delta z [\kappa(\nu_1) - \kappa(\nu_2)] + \tilde{F}(x, \nu_1, z) - \tilde{F}(x, \nu_2, z), \quad (14.11)$$

with  $\Delta z = z_2 - z_1$  and the function  $\tilde{F}$

$$\tilde{F}(x, \nu_i, z) = \ln \frac{1 - x(z_1) + x(z_1)\tilde{L}(\nu_i, z_1)}{1 - x(z_2) + x(z_2)\tilde{L}(\nu_i, z_2)} \quad (14.12)$$

which accounts for the aerosol structure of the atmosphere by  $x$  and the change in the backscattered spectrum by the normalized spectrum function

$$\tilde{L}(\nu_i, z_j) = \frac{(L(z_j) * rbs)(\nu_i)}{L(\nu_i, z_j)} = \frac{\int_{-\infty}^{\infty} L(\nu', z_j) \cdot rbs(\nu_i - \nu') d\nu'}{L(\nu_i, z_j)}. \quad (14.13)$$

It should be mentioned that for the derivation of Eq. (14.11) the usual DIAL assumptions are made that (1) the overlap function  $O(z)$  is the same for both wavelengths and (2) the wavelength dependence of the extinction and backscatter coefficients  $\alpha$  and  $\beta$  of the unperturbed atmosphere (i.e., without the gas of interest) are negligible over the wavenumber interval  $\nu_1, \nu_2$ .

In the hypothetical case  $x(z_1) = x(z_2) = 0$ , i.e., pure particle scattering with no spectral broadening, the function  $\tilde{F}$  vanishes and Eq. (14.11) turns into to the familiar DIAL approximation

$$Q(z) = 2 \cdot \Delta z [\kappa(\nu_1) - \kappa(\nu_2)]. \quad (14.14)$$

In reality both scattering processes contribute to the return signal. Two extreme cases can be distinguished: homogeneous and inhomogeneous scattering. In the first case the maximum contribution of  $\tilde{F}$  in Eq. (14.11) occurs for  $x(z_1) = x(z_2) = 0.5$  [11]. From Eq. (14.12) it follows that

$$\tilde{F}_{\text{hom}}^{\text{max}}(\nu, z) = \ln \frac{1 + \tilde{L}(\nu, z_1)}{1 + \tilde{L}(\nu, z_2)}. \quad (14.15)$$



For the second, the inhomogeneous case  $x(z_1) = 1$ ,  $x(z_2) = 0$  or vice versa, it is given by

$$\tilde{F}_{\text{inh}}^{\text{max}}(\nu, z) = \ln[\tilde{L}(\nu, z_1)]. \quad (14.16)$$

It is known that the latter case ( $\equiv$  gradients of aerosol concentration) causes the dominant error which can range up to 100% for water vapor [10] and up to 10 K for temperature measurements [11].

In order to correct for this effect, i.e., to calculate  $x(z)$  the backscatter coefficient must be known with good accuracy ( $\approx 5\%$  for 1 K [8]). Normally no independent information, e.g., from a Raman lidar, is available. Thus the inversion algorithm by Klett [12] and Fernald [13] is applied to the off-resonance signal. This method leads to the differential equation—in the notation of Bissonnette [14]—

$$\frac{d\mathcal{P}}{dz} = \frac{1}{\beta(z)} \frac{d\beta(z)}{dz} - 2 \cdot \alpha(z). \quad (14.17)$$

$\mathcal{P}$  is the measured range-corrected “off-resonance” signal from which two unknowns ( $\alpha, \beta$ ) must be derived. This is of course not possible without making assumptions on the relationship between  $\beta$  and  $\alpha$ . A power law of the kind  $\alpha^\epsilon = S^*(z) \cdot \beta$  is often assumed. Although with this relation Eq. (14.17) becomes a differential equation of Bernoulli type with known solution, the underdetermination still persists since  $S^*(z)$  is not known either. To circumvent or at least to reduce the influence of these uncertainties the BELINDA scheme has been suggested [15].

### 14.2.3 BELINDA

#### Eliminating the Influence of Broadening Processes

The idea of BELINDA is to get rid of the  $\tilde{F}$  terms in Eq. (14.11). Assume a broad laser emission spectrum of Gaussian shape with approximately twice the half width of the absorption feature is transmitted into the atmosphere. The resulting backscattered spectrum, calculated with the absorption line parameters of water vapor in the vicinity of 720 nm, is sketched in Fig. 14.1. For this computation, a height of 1000 m, standard atmospheric conditions, and pure particle scattering are assumed. The corresponding spectrum for pure molecular scattering, i.e., with Doppler line broadening, is shown in Fig. 14.2. Four points of intersection of the two spectra are identified, two close to the absorption maximum (central dip) and two in the wings of the absorption feature. At these frequencies

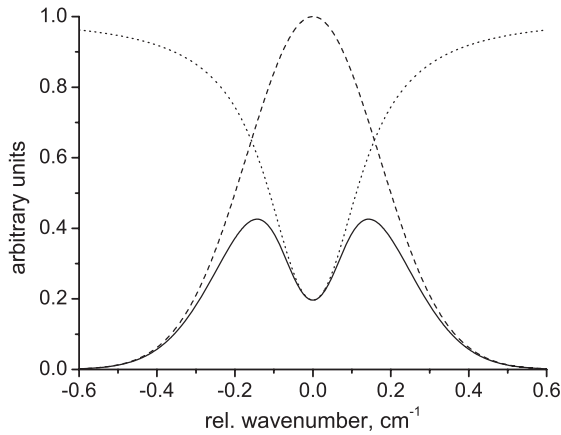


**Fig. 14.1.** Backscattered spectrum (solid line) of a broad laser emission line (dashed) attenuated by the absorption feature (dotted). No broadening assumed.

the convolution integral in Eq. (14.13) is equal to the denominator, and  $\tilde{L}$  is unity. Insertion into Eq. (14.12) gives  $\tilde{F} = \ln(1/1) \equiv 0$  and therefore  $Q(z)$  in Eq. (14.11) becomes independent of the backscatter properties of the atmosphere.

It is true that the independence of this measuring scheme on the scattering process holds, strictly speaking, only for this particular height.

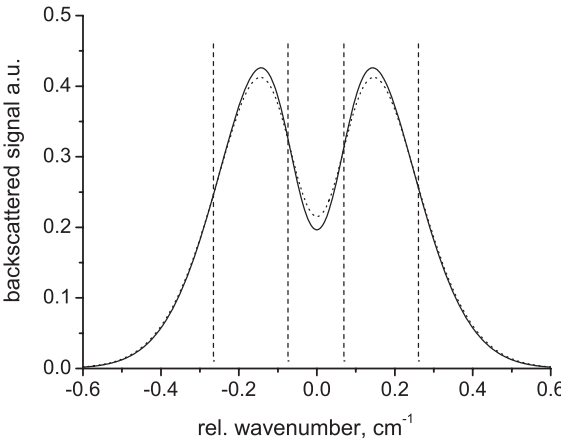


**Fig. 14.2.** Normalized unbrodened (as in Fig. 14.1) and broadened backscatter spectra (solid and dotted line, respectively). The intersections of the two are marked by dashed vertical lines.

To see what happens throughout the atmosphere a somewhat more detailed analysis is required.

## Simulations

The following calculations were made with the parameters [16] of the water-vapor absorption line at 728.94 nm ( $\nu_0 = 13718.58 \text{ cm}^{-1}$ ) suitable for measurements in the planetary boundary layer. This does not mean any loss of generality because the results scale to good approximation with the ratio of the laser to the absorption feature linewidth. Standard atmospheric condition, i.e.,  $T_0 = 20^\circ\text{C}$ ,  $p_0 = 1013 \text{ hPa}$ , and a relative humidity of 50% are assumed.

Simulations were carried out to show the effect of height on the backscattered signal as a function of shift of the wavenumber relative to absorption line center ( $\Delta\nu = \nu - \nu_0$ ) for the inhomogeneous [ $\tilde{F}_{\text{inh}}^{\text{max}}$ , Eq. (14.16)] and, for the sake of completeness, the homogeneous [ $\tilde{F}_{\text{hom}}^{\text{max}}$ , Eq. (14.15)] case also. Results are plotted in Fig. 14.3.

As already stated, the error due to homogeneous scattering cannot be distinguished in this representation and is thus negligible. As one can also see the intersection points, i.e., the wavenumbers for which  $\tilde{F}_{\text{inh}}^{\text{max}} = 0$ , shift with height toward larger wavenumber differences. In order to minimize the contribution of  $\tilde{F}$  in Eq. (14.11) the selection of the “on-resonance” and “off-resonance” wavenumber should be done in

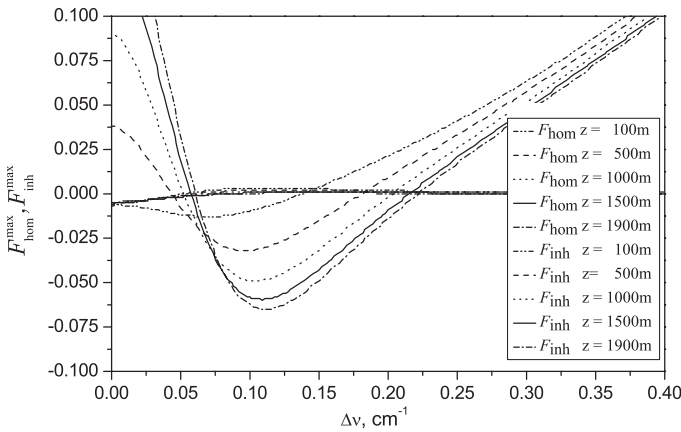


**Fig. 14.3.** Maximum error due to homogeneous and inhomogeneous scattering for different heights, laser linewidth (HWHM)  $0.2 \text{ cm}^{-1}$ ,  $\Delta z = 100 \text{ m}$ .

such a way that

$$\tilde{E}(\nu_1, \nu_2) = \sum_{z_j=z_{\min}}^{z_{\max}} \left| \tilde{F}_{\text{inh}}^{\max}(\nu_1, z_j) - \tilde{F}_{\text{inh}}^{\max}(\nu_2, z_j) \right| \quad (14.18)$$

gets minimal. This is the accumulated error over the whole measurement range. A slightly different solution is obtained by calculating the relative error for each range cell [17]

$$\frac{d\kappa}{\kappa}(\nu_1, \nu_2, z_j) = \frac{\left| \tilde{F}_{\text{inh}}^{\max}(\nu_1, z_j) - \tilde{F}_{\text{inh}}^{\max}(\nu_2, z_j) \right|}{2 \cdot \Delta z \cdot [\kappa(\nu_1, z_j) - \kappa(\nu_2, z_j)]} \quad (14.19)$$

from which the average error for the whole measurement range is computed as

$$\langle \delta\kappa \rangle = \frac{\Delta z}{z_{\max} - z_{\min}} \sum_{z_j=z_{\min}}^{z_{\max}} \frac{d\kappa}{\kappa}(\nu_1, \nu_2, z_j). \quad (14.20)$$

The values of  $\langle \delta\kappa \rangle$  obtained by varying, in Eq. (14.19),  $\Delta\nu_1$  and  $\Delta\nu_2$  over suitable ranges are shown in Fig. 14.4. As expected, the result is symmetric in  $\nu_1, \nu_2$ . The minima form a shallow valley of hyperbolic shape, i.e.,  $\nu_1 \cdot \nu_2 \approx \text{const}$ . The absolute minimum is found at  $\nu_1, \nu_2 = 0.11 \text{ cm}^{-1}$  which is of course no suitable solution since the differential absorption coefficient is zero for  $\nu_1 = \nu_2$ . Therefore a figure-of-merit function (FOM) is defined as

$$\text{FOM} = \langle \delta\kappa \rangle \cdot 1/\kappa. \quad (14.21)$$

Figure 14.5 shows the computational result. Searching again for the minimum yields the indicated values for the “on” and “off”-wavenumber difference. The corresponding averaged error (see Fig. 14.4) is  $\approx 17\%$  which is only a slight increase compared to the absolute minimum of  $\approx 12\%$ . The range-resolved error and optical thickness are plotted in Fig. 14.6. Whereas the optical thickness varies slowly with range according to the assumed water-vapor profile, the error due to inhomogeneous scattering ranges between 30% at the lowest and highest range gates and zero at 1300 m and 1450 m, depending on the laser linewidth.

For a typically evolved planetary boundary layer in the mid-latitudes steep gradients in aerosol content are common between 1000 and 1500 m. It is a desirable feature that for this height range the error due to inhomogeneous scattering is minimal and below 10%, which is normally

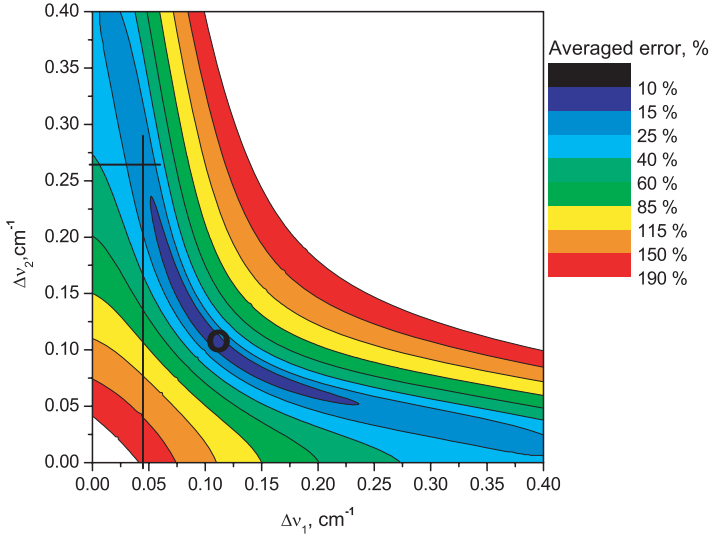


**Fig. 14.4.** Average error according to Eq. (14.20) as a function of the spacing between the “on-” and “off-resonance” receiver transmission maximum and absorption line center. Note the progressive scaling of the color ramp. The minimum is indicated by the black circle and the optimum setting (see below) by the black cross. Laser linewidth (HWHM)  $0.2 \text{ cm}^{-1}$ ,  $\Delta z = 100 \text{ m}$ .

acceptable, particularly as the aerosol content does not jump from zero to one as it was assumed for these calculations. It is noteworthy that this result is obtained by the BELINDA approach without any assumptions on the backscatter properties of the atmosphere.

One of the next steps is to expand the figure-of-merit function [Eq. (14.21)] in such a way as to account for further system parameters of which the laser bandwidth is probably the most important. Work in this direction is in progress.

#### 14.2.4 Practical Considerations

From a practical point of view an important difference between BELINDA and conventional DIAL is the shift of the frequency selection, wavelength narrowing and most of the wavelength stabilization from the transmitting to the receiving end of the system with important consequences particularly for those applications in which single, narrow, well-separated lines of a gas of interest are used. The near infrared between 700 and 900 nm is such a region.

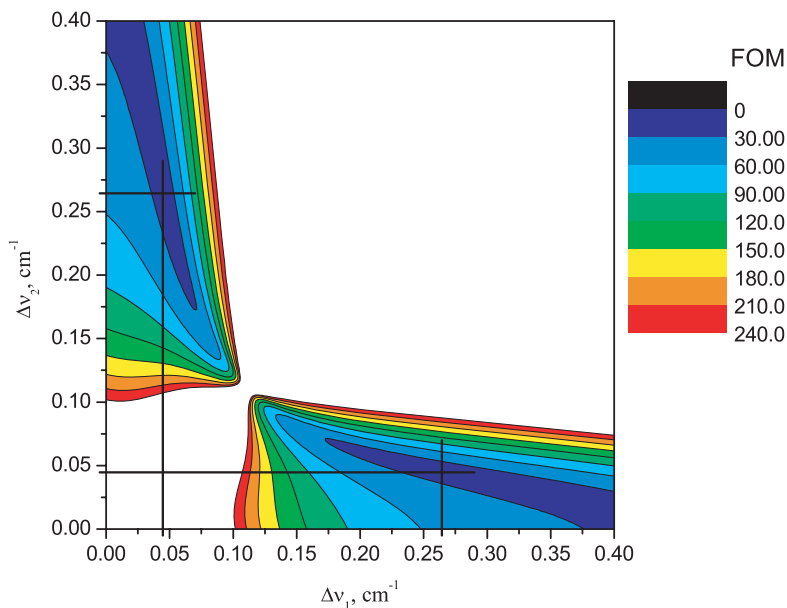


**Fig. 14.5.** Figure-of-merit values (a.u.) according to Eq. (14.21) in dependence of the spacing for on- and off-line separation, respectively. The minimum is located at  $\Delta\nu_1 = 0.045 \text{ cm}^{-1}$  and  $\Delta\nu_2 = 0.265 \text{ cm}^{-1}$  and vice versa as indicated by the black cross. Laser linewidth (HWHM)  $0.2 \text{ cm}^{-1}$ ,  $\Delta z = 100 \text{ m}$ .

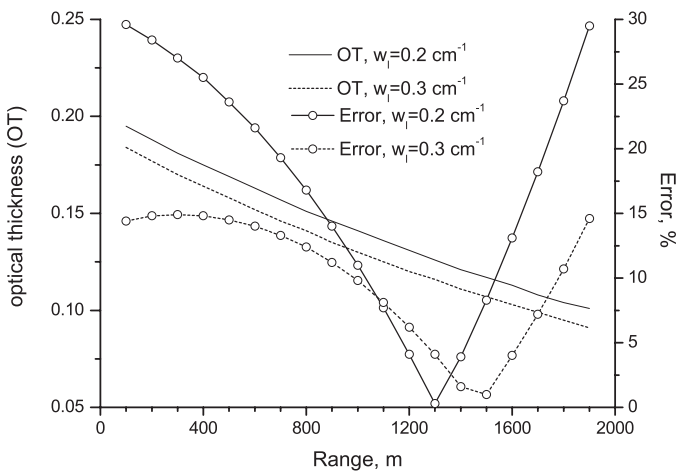


**Fig. 14.6.** Range resolved optical thickness and error due to inhomogeneous scattering for each range cell ( $\Delta z = 100 \text{ m}$ ) for two values of the laser linewidth (HWHM).

The main result, as outlined above and confirmed by measurements [17], is the fact that BELINDA results are nearly independent of the atmospheric aerosol distribution. As was shown in Chapters 7 and 8 of this book, great care must be taken in conventional DIALs that gradients of aerosol properties do not fake gas concentrations even if the on- and off-resonance wavelengths are quite close together. This inherent sensitivity to aerosols is drastically reduced by the BELINDA principle.

### Transmitter

Whereas common DIAL systems use two lasers or one laser that is switched between at least two wavelengths, for the BELINDA approach only one laser and no wavelength switching is necessary. Apart from the obvious savings in weight, space, cost, and beam combining equipment with respect to the two-laser systems and the freedom from limitations to low-pulse-repetition frequencies and the resulting “non-frozen-atmosphere-problem” of the switched systems, the problem of ensuring that pulses of both wavelengths overlap sufficiently well in space is inherently nonexistent. This means that even the short-range returns for which the overlap function  $O(z)$  is not unity can be evaluated. This is practically impossible in classical DIAL.

Laser wavelength adjustment and stability are also less critical in BELINDA than in conventional DIAL. As deviations from nominal line position and width have much less effect on the result, no complicated feedback stabilization system is necessary. For BELINDA the laser linewidth must be  $\approx 0.2 \text{ cm}^{-1}$  which is 15–20 times broader than for conventional DIAL. This broadband emission cannot be obtained with a single laser mode which is of the order of  $0.001 \text{ cm}^{-1}$ . Thus the wavelength-selective elements of the laser are chosen in such a way that sufficient laser modes can oscillate to form the required spectrum. The wide spectrum does not have to be covered by each individual laser pulse, only over the integration time needed for one profile. The reason for this is that (i) the DIAL approximation, Eq. (14.11), is independent of the transmitted energy, (ii) the convolution integral, Eq. (14.8), can be thought of as a summation over an infinite number of broadened laser modes, and (iii) interferometers as used in the receiver (see below) work even with one photon.

Narrow-output lasers, particularly when tunable over a wide range of frequencies, may show a “socket” of amplified spontaneous emission (ASE) on which the narrow emission line is superimposed. This undesirable light is efficiently rejected by the interferometers of the

receiver leading to a much smaller ASE sensitivity for the BELINDA approach.

Finally, the modeling of the performance shows that inaccuracy and shifts of the laser center frequency as large as  $\pm 0.01 \text{ cm}^{-1}$  cause a concentration error of only 0.5% if a receiver is used that is conceived along the lines sketched below.

## Receiver

Theoretically two narrow-bandpass filters, each adjusted to transmit the wavelength where the unbroadened and broadened backscatter spectrum intersect, respectively (Fig. 14.2), would be sufficient. Then wavenumber setting and stability of the filters, particularly of that closer to the center of the absorption line (the “on-resonance” filter), would have to be better than  $0.01 \text{ cm}^{-1}$  for a relative change of 10% of the absorption coefficient. The situation gets much more relaxed, by a factor of 10, if the backscattered signal is detected with four instead of two transmission peaks, symmetrical on either side of the absorption line. This is due to the fact that the absorption coefficient in the vicinity of  $0.05 \text{ cm}^{-1}$  from line center varies approximately linear with wavelength. Thus a misalignment leads to a decrease on one side which is compensated by an increase on the opposite side. This compensation holds also for the  $\tilde{F}$  terms in Eq. (14.11).

The same argument applies to the “off-resonance” filter; it proved advantageous to use neighboring transmission modes of a normal Fabry–Perot interferometer (FPI) positioned symmetrically at a distance of  $0.26 \text{ cm}^{-1}$  (see Fig. 14.5) at either side of the absorption line. The other transmission modes are so far off the laser line that normal daylight blocking is sufficient to ensure an adequate signal-to-noise ratio.

This condition is not met for the “on-resonance” filter if a conventional FPI is used. If, however, a double-cavity étalon (DCE) [18] consisting of three equally spaced parallel mirrors is utilized, each transmission peak splits into two. The amount of this splitting is conveniently controlled by the reflectivity of the inner mirror whereas the free spectral range, i.e., the spectral separation between adjacent peak pairs, is governed by the spacing of the mirrors. Physically the DCE consists of two plane-parallel flats dielectrically coated prior to optically contacting.

Figure 14.7 shows the transmission features of a usual FPI (“off-resonance” signals) and a DCE (“on-resonance” signals). Much like the FPI, the DCE can also be adjusted to the desired wavelength by tilting. All



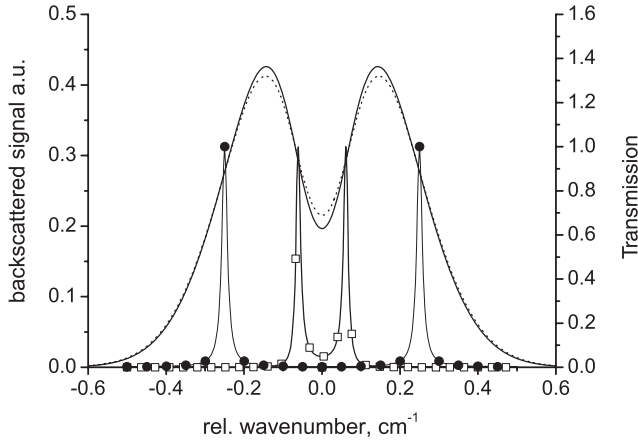


**Fig. 14.7.** Measurement principle of the BELINDA approach. Backscatter spectra as of Fig. 14.2 and transmission of the double-cavity étalon (open squares) and a Fabry–Perot interferometer (solid circles).

that is needed to maintain the required stable transmission wavelengths is a housing stabilized to  $\pm 0.1$  K which then corresponds to a measuring error of 0.5%—slightly dependent on the interferometer material.

The filters reject  $\approx 90\%$  of the backscattered light. The actual efficiency is even lower than the modeled value of 10%, approaching 7% in practical cases. This drawback, however, is more than balanced by the remaining features. Table 14.1 summarizes the quintessential

**Table 14.1.** Comparison of main characteristics of the BELINDA with those of a usual DIAL setup

| Property                                       | BELINDA                       | DIAL                                              |
|------------------------------------------------|-------------------------------|---------------------------------------------------|
| Aerosol correction                             | Not mandatory                 | Required                                          |
| Data evaluation where $O(z) \leq 1^{\S}$       | No restriction                | Difficult                                         |
| Sensitivity to ASE*                            | Low                           | High                                              |
| On/off pulse delay $\Delta t$                  | Not applicable                | $200 \mu\text{s} \leq \Delta t \leq 1 \text{ ms}$ |
| Number of laser(s)                             | 1                             | 2, or 1 with switching                            |
| Number of optical axes                         | 2 (laser + telescope)         | 3 (2 lasers + telescope)                          |
| Usable laser power                             | 7%                            | 100%                                              |
| <b>Laser spectral requirements<sup>†</sup></b> |                               |                                                   |
| Bandwidth (HWHM)                               | $0.200/0.100 \text{ cm}^{-1}$ | $0.013/0.005 \text{ cm}^{-1}$                     |
| Line center accuracy                           | $0.020/0.010 \text{ cm}^{-1}$ | $0.005/0.002 \text{ cm}^{-1}$                     |

<sup>\S</sup> $O(z)$ : overlap integral, \*ASE: amplified spontaneous emission, <sup>†</sup>for 1% systematic error on water vapor/temperature

characteristics of the BELINDA principle. Whereas the first four lines show properties that cannot or can only to a certain degree be influenced by the experimenter, the remaining five properties are more technical in nature.

## 14.3 White-Light Femtosecond Lidar

### 14.3.1 Non-Linear Propagation of Terawatt Pulses

High-power laser pulses propagating in transparent media undergo a number of nonlinear effects. Nonlinear self-action leads to strong evolutions of the spatial (self-focusing [19, 20], self-guiding [21], self-reflection [22]), spectral (four-wave mixing [23], self-phase modulation [24–26]) as well as temporal (self-steepening [27], pulse splitting [28]) characteristics of the pulse. The propagation medium is also affected, as it is partially ionized by the propagating laser beam [29–32].

These phenomena have been extensively studied since the early 1970s, from the theoretical as well as from the experimental point of view. It was only in 1985, however, that the development of the chirped-pulse amplification (CPA) technique [33, 34] permitted to produce ultrafast laser pulses, to reach intensities as high as  $10^{20}$  W/cm<sup>2</sup> and hence to observe highly nonlinear propagation even in slightly nonlinear media such as atmospheric-pressure gases. We will focus here on nonlinear propagation in air and on processes related to coherent white-light generation and filamentation.

### Kerr Self-Focusing

For high intensities  $I$ , the (real part of the) refractive index  $n$  of the air is modified by the Kerr effect [19, 20], becoming

$$n(I) = n_0 + n_2 \cdot I \quad (14.22)$$

where  $n_0 = 1.000293$  is the refractive index for 800 nm wavelength at 0 °C and atmospheric pressure and  $n_2 = 3 \times 10^{19}$  cm<sup>2</sup>/W is the nonlinear refractive index of the air. As the intensity in a cross section of the laser beam is not uniform, the refractive index increases more strongly in the center of the beam than on the edge (Fig. 14.8a).

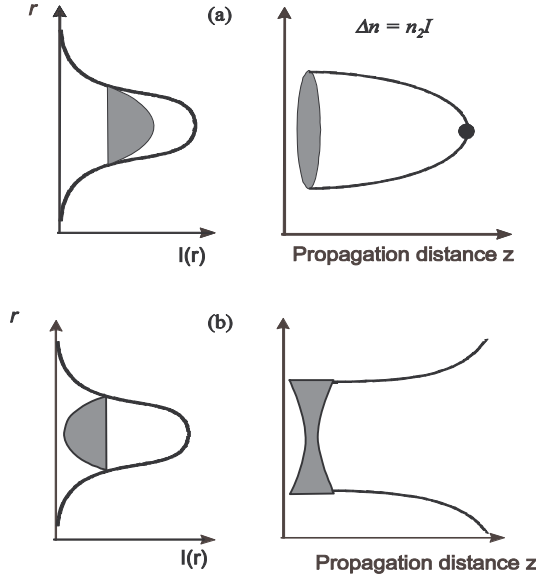


**Fig. 14.8.** Kerr self-focusing (a) and plasma-induced self-defocusing (b) of the laser beam. The radial intensity profile yields refractive index gradients that act as a positive (a) or negative (b) lens as the laser beam propagates.

This induces a radial refractive index gradient equivalent to a converging lens (called “Kerr lens”) of focal length  $f(I)$ . The beam is focused by this lens, which leads to an intensity increase, which leads in turn to a lens of shorter focal length, and so on until the whole beam collapses on itself. Kerr self-focusing should therefore prevent propagation of high-power lasers in air.

The Kerr effect becomes significant when self-focusing gets larger than natural diffraction, i.e., when the pulse power exceeds a critical power

$$P_{\text{crit}} = \frac{\lambda^2}{4\pi \cdot n_2}. \quad (14.23)$$

It should be pointed out that this is a critical *power* rather than a critical intensity. For a titanium-sapphire laser ( $\lambda = 800 \text{ nm}$ ) in air,  $P_{\text{crit}} \sim 2 \text{ GW}$  (a more detailed treatment shows that in case of pulses shorter than 100 fs,  $P_{\text{crit}} \sim 6 \text{ GW}$ ). Conversely, the distance at which the beam is focused is related to the successive focal lengths  $f(I)$  and is a function of the initial *intensity*.

## Multiphoton Ionization and Plasma Generation

If the laser pulse intensity reaches  $10^{13}$ – $10^{14}$  W/cm<sup>2</sup>, higher-order nonlinear processes occur such as multiphoton ionization (MPI). At 800 nm, 8–10 photons are needed to ionize N<sub>2</sub> and O<sub>2</sub> molecules and give rise to the formation of a plasma [35]. The ionization process can involve tunneling as well, because of the very high electric field carried by the laser pulse. However, following Keldysh's theory [36], MPI dominates for intensities  $<10^{14}$  W/cm<sup>2</sup>. In contrast to longer pulses, fs pulses combine high ionization efficiency due to their very high intensity with a limited overall energy, so that the generated electron densities  $\rho$  of  $10^{16}$ – $10^{17}$  cm<sup>-3</sup> are far from saturation. Losses by inverse bremsstrahlung are therefore negligible, in contrast with ns (or longer)-pulse-laser generated plasma. However, the electron density induces a negative variation of the refractive index and, because of the radial intensity profile of the laser beam, a negative refractive index gradient. This acts as a negative lens which defocuses the laser beam, as schematically shown in Fig. 14.8b.

## Filamentation of High-Power Laser Beams

Kerr self-focusing and plasma defocusing should thus prevent long-distance propagation of high-power laser beams. However, a remarkable behavior is observed in air, where both effects exactly compensate and give rise to self-guided quasi-solitonic [37] propagation. The laser beam is first self-focused by the Kerr effect. This focusing then increases the beam intensity and generates a plasma by MPI, which in turns defocuses the beam. The intensity then decreases and plasma generation stops, which allows Kerr re-focusing to take over again. This dynamic balance between Kerr effect and plasma generation leads to the formation of stable structures called “filaments” (Fig. 14.9). Light filaments in air were first observed by Braun et al. [21], who discovered that mirrors could be damaged by high-power ultrashort laser pulses even at large distance from the laser source. These light filaments have remarkable properties. In particular, they can propagate over several hundreds of meters, although their diameter is only 100–200  $\mu$ m (thus widely beating the usual diffraction limits), and have almost constant values of intensity (typically  $10^{14}$  W/cm<sup>2</sup>), energy (a few mJ), diameter, and average electron density ( $10^{16}$ – $10^{17}$  cm<sup>-3</sup>).

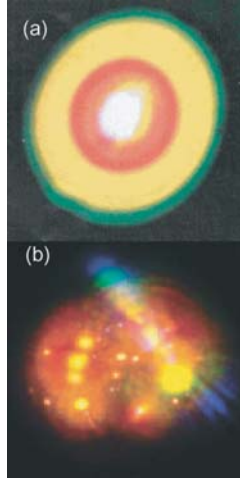


**Fig. 14.9.** Pictures of laser beam cross-sections in case of a 50 GW (a) and a 5 TW (b) laser, showing filamentation. For the lower beam power, a single filament is produced (a), while for the TW laser the beam breaks up in a large number of filaments (b). Notice the conical emission associated with each filament, and the coherence between different filaments that leads to interference patterns. Pictures (a) and (b) are not at the same scale.

More precisely, the laser pulse propagation is governed by the Maxwell wave equation

$$\nabla^2 E - \frac{1}{c^2} \cdot \frac{\partial^2 E}{\partial t^2} = \mu_0 \cdot \sigma \cdot \frac{\partial E}{\partial t} + \mu_0 \cdot \frac{\partial^2 P}{\partial t^2}. \quad (14.24)$$

$E$  is the magnitude of the electric field vector,  $\sigma$  is the conductivity which accounts for losses, and  $P$  is the polarization of the medium.  $\mu_0$  is the magnetic permeability of vacuum and  $c$  the speed of light. In contrast with the linear wave propagation equation,  $P$  now contains a self-induced non-linear contribution corresponding to Kerr focusing and plasma generation:

$$P = P_L + P_{NL} = \varepsilon_0 \cdot (\chi_L + \chi_{NL}) \cdot E. \quad (14.25)$$

Here  $\chi_L$  and  $\chi_{NL}$  are the linear and nonlinear susceptibilities, respectively, and  $\varepsilon_0$  is the permittivity of vacuum. Considering a radially symmetric pulse propagating along the  $z$  axis in a reference frame moving at the group velocity  $v_g$  yields the nonlinear Schrödinger equation

(NLSE) [37]

$$\nabla_{\perp}^2 \varepsilon + 2i \left( k \frac{\partial \varepsilon}{\partial z} \right) + 2k^2 n_2 \cdot |\varepsilon|^2 \cdot \varepsilon - k^2 \frac{\rho}{\rho_c} \cdot \varepsilon = 0 \quad (14.26)$$

where  $\varepsilon = \varepsilon(r, z, t)$  is the pulse envelope of the electric field,  $\rho_c$  the critical electron density ( $1.8 \times 10^{21} \text{ cm}^{-3}$  at 800 nm [37]), and  $k = 2\pi/\lambda$ .  $\varepsilon$  is assumed to vary slowly as compared to the carrier oscillation and to have a smooth radial decrease. In this first-order treatment, group velocity dispersion (GVD) and losses due to multiphoton and plasma absorption are neglected ( $\sigma = 0$ ). In Eq. (14.26), the Laplacian models wave diffraction in the transverse plane, while the two last terms are the non-linear contributions: Kerr focusing and plasma defocusing (note the opposite signs). The electron density  $\rho(r, z, t)$  is computed using the rate equation (14.27) in a self-consistent way with Eq. (14.26):

$$\frac{\partial \rho}{\partial t} - \gamma |\varepsilon|^{2\alpha} (\rho_n - \rho) = 0. \quad (14.27)$$

Here  $\rho_n$  is the neutral-molecule concentration in air,  $\gamma$  the MPI efficiency and  $\alpha$  the number of photons needed to ionize an air molecule (typically  $\alpha = 10$  [35]). Solving the NLSE (numerically) leads to the evolution of the pulse intensity  $I = |\varepsilon|^2$  as a function of propagation distance, as shown schematically in Fig. 14.10. Initial Kerr-lens self-focusing and subsequent stabilization by the MPI-generated plasma are well reproduced by these simulations. Notice that the filamentary structure of the beam, although only 100  $\mu\text{m}$  in diameter, is sustained over 60 m. Numerical instability related to the high non-linearity of the NLSE prevents simulations over longer distances.

For laser powers  $P \gg P_{\text{crit}}$  the beam breaks up into several localized filaments. The intensity in each filament is indeed clamped at

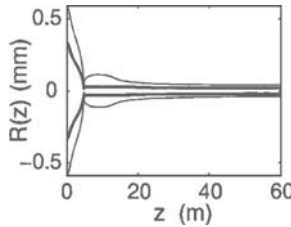


**Fig. 14.10.** Numerical simulations showing the process of filamentation in air [37]. After an initial collapse due to Kerr self-focusing, the beam stabilizes in a filamentary structure of typically 100  $\mu\text{m}$  in diameter that propagates over very long distances (here 60 m).

$10^{13}$ – $10^{14}$  W/cm<sup>2</sup> corresponding to a few mJ, so that an increase in power leads to the formation of more filaments. Figure 14.9 shows a cross section of laser beams undergoing monofilamentation (Fig. 14.9a, 5 mJ) and multifilamentation (Fig. 14.9b, 400 mJ). The stability of this quasi-solitonic structure is remarkable: filaments have been observed to propagate over more than 300 m but no direct measurements could be made at longer distances yet because of experimental constraints.

Many theoretical studies have been carried out to simulate the non-linear propagation of high-power laser beams, both in the monofilamentation [38–46] and in the multifilamentation [47–49] regimes.

### White-Light Generation and Self-Phase Modulation (SPM)

The spectral content of the emitted light is of particular importance for lidar applications. Nonlinear propagation of high-intensity laser pulses not only provides self-guiding of the light but also an extraordinarily broad continuum extending from the UV to the IR. This supercontinuum is generated by self-phase modulation as the high-intensity pulse propagates. As depicted above, the Kerr effect leads, because of the spatial intensity gradient, to self-focusing of the laser beam. However, the intensity also varies with time, and the instantaneous refractive index of the air is modified according to

$$n(t) = n_0 + n_2 \cdot I(t). \quad (14.28)$$

This gives rise to a time-dependent phase shift  $d\phi = -n_2 I(t) \cdot \omega_0 z / c$  (where  $\omega_0$  is the carrier frequency), which generates new frequencies  $\omega = \omega_0 + d\phi/dt$  in the spectrum. The smooth temporal envelope of the pulse induces thus a strong spectral broadening of the pulse about  $\omega_0$ . Figure 14.11 shows the spectrum emitted by filaments that were created by the propagation of a 2-TW pulse in the laboratory. The supercontinuum extends from over 4  $\mu$ m down to 400 nm. Recent measurements in air showed an extraordinary UV extension to 230 nm [50] due to efficient third-harmonic generation (THG) and frequency mixing [51, 52]. It thus covers absorption bands of many trace gases in the atmosphere such as methane, volatile organic compounds (VOCs), aromatics, CO<sub>2</sub>, NO<sub>2</sub>, H<sub>2</sub>O, SO<sub>2</sub>, and ozone with promising new, multispectral lidar measurements of these gases.

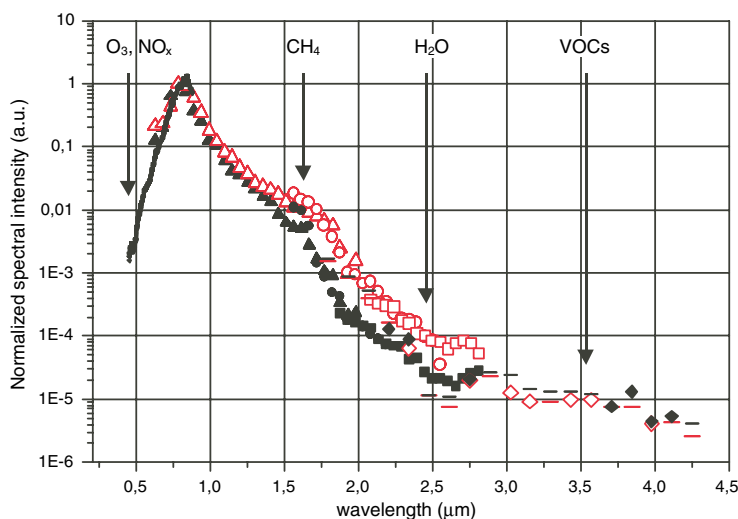


**Fig. 14.11.** Laboratory measurements of the supercontinuum spectrum generated by self-phase-modulation (SPM). The obtained “white-light laser” covers a spectral range from 400 nm to 4  $\mu\text{m}$  [56]. Some typical atmospheric trace gas absorption regions are indicated. Recent measurements in air showed a dramatic UV extension down to 230 nm. Different colors and different symbols indicate different chirp settings.

### Angular Distribution of the Supercontinuum Emission

Most of the filamentation studies showed that white light was generated in the filamentary structure, and, due to coupling with the plasma, leaking into the forward direction as a narrow cone. This “conical emission” (Fig. 14.9a), [53, 54] with the longer wavelengths in the center to the shorter wavelengths at the edge, extends over a typical half-angle of  $0.12^\circ$ .

However, a more important aspect for lidar applications is the angular distribution of the white light continuum in the near-backward direction. In the first fs lidar experiments already, a pronounced backscattering component of the emitted white light was observed. For this reason angle-resolved scattering experiments were carried out. The emission close to the backward direction of the supercontinuum from light filaments was found to be significantly enhanced as compared to linear Rayleigh–Mie scattering [55]. Figure 14.12 shows a comparison of the linearly backscattered light (Rayleigh–Mie) from a weak laser beam with the non-linear emission from a filament, for both s (left part) and p (right part) polarizations. At  $179^\circ$  the backward enhancement amounts to almost an order of magnitude. An even greater enhancement is expected at



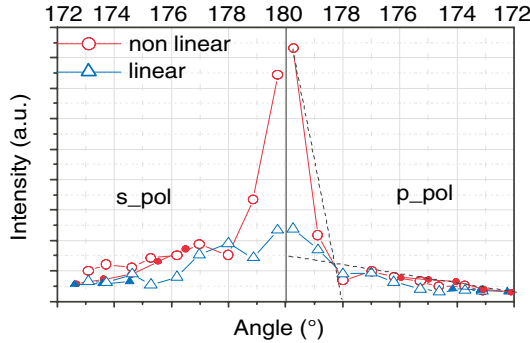


**Fig. 14.12.** Angular dependence of the white-light intensity emitted by a filament. A strong backward enhancement is observed for both s (left) and p polarization (right). The nonlinear emission (circles) is compared to elastic scattering (triangles) from a low-energy laser beam in the same wavelength region (400–450 nm).

180°, but could not be observed because of the limitations of the experimental apparatus. The enhancement may qualitatively be attributed to a co-propagating, self-generated longitudinal index gradient due to plasma generation, inducing backreflection and stimulated Raman scattering. Combined with self guiding which drastically reduces beam divergence, this aspect is extremely important for lidar experiments: a large fraction of the white light is thus collected by the lidar receiver, unlike radiation from conventional elastic scattering of white light emitted, e.g., by flashlamp-based lidars [57].

To summarize, nonlinear propagation of TW laser pulses exhibits several unique properties for multispectral lidar measurements, namely, extremely broadband coherent light emission (“white light laser”) confined in a self-guided beam and back-reflected to the emitter as the laser pulse propagates.

### 14.3.2 The TERAMOBILE Project

Based on the first experiments [2, 3] with fs white light in a lidar arrangement, a large-frame French–German project called TERAMOBILE (for “Terawatt laser in a mobile system”) was launched in 1999. Its aim was to design and build the first mobile TW-laser-based lidar system, investigate fundamental processes like long-range propagation and filamentation, and develop new possibilities of sounding the atmosphere.

The Teramobile system [58, 50] consists of a femtosecond-terawatt laser and a multispectral lidar detection system integrated in an air-conditioned container. Mobility is a crucial aspect, but also a strong constraint. Particular care has therefore been given to the design of the 20-ft mobile container laboratory (Fig. 14.13).

It is equipped with windows in the roof and in the wall to permit horizontal and vertical measurements. The system needs only cooling water and power as external supplies. For full stand-alone operation a separate mobile unit is also available to provide cooling and electric power from a diesel generator.

The heart of the system is a TW-fs chirped-pulse amplification (CPA) laser system provided by Thales Laser (formerly BMI-Thomson). The fs pulses (a few nJ, 60 fs) are generated by a Kerr-lens mode-locked Ti:sapphire oscillator pumped by a cw Nd:YLF laser. The pulses are then stretched to about 500 ps using a grating arrangement in order to prevent damage in the amplifier chain. Amplification is carried out with a regenerative amplifier, a multipass preamplifier, and a final multipass amplifier pumped by two Nd:YAG lasers of 1 J energy each at 10 Hz. The amplified pulse is then recompressed by a second pair of gratings to

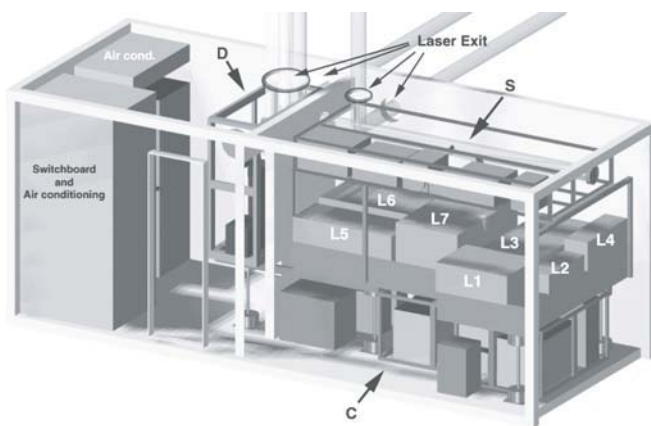


**Fig. 14.13.** The TERAMOBILE lidar system. The system is split into: (1) a laser room that contains the CPA-femtosecond TW laser (oscillator (L1), stretcher (L2), regenerative amplifier, multipass preamplifier (L3), YAG pump laser (L4), main amplifier (L5), YAG pump lasers (L6), compressor (L7)), YAGs power supplies and heat exchanger (C), and beam expanding optics (S) and (2) a control room that contains the lidar receiver (D) and the signal processing electronics. Both vertical and horizontal measurement configurations are possible.

60 fs pulse length with correspondingly high intensity. A key feature of the grating compressor is that a chirp can be applied to the output laser pulses. Negative chirping, i.e., a regime in which the shorter wavelength components precede the longer wavelength components, is an efficient means for the compensation of group velocity dispersion in the infrared (see Subsection 14.3.3). The pulse duration, and thus the peak intensity, can also be modified in this way.

Because of eye-safety considerations, the femtosecond lidar is assisted by a stand-alone low-power eyesafe lidar [59] with wide field of view that detects any air vehicle entering the measurement region and shuts down the fs lidar.

The most important technical data of the system are presented in Table 14.2.

### 14.3.3 White-Light Femtosecond Lidar Measurements

Since filamentation counteracts diffraction over long distances, it allows to deliver high laser intensities at high altitudes and over long ranges. This contrasts with linear propagation, in which the intensity always decreases as the beam propagates away from the source, unless focusing optics such as large-aperture telescopes with adaptive optics are used to generate focal lengths of the order of hundreds of meters.

**Table 14.2.** Technical data of Teramobile lidar

|                                      |                             |
|--------------------------------------|-----------------------------|
| Laser:                               |                             |
| Pulse duration                       | 60 fs FWHM                  |
| Pulse energy                         | 330 mJ                      |
| Peak power                           | 5.5 TW                      |
| Pulse-to-pulse stability             | 2% RMS                      |
| Bandwidth                            | 16 nm FWHM                  |
| Beam transmitter telescope:          |                             |
| Input pulse diameter                 | 50 mm                       |
| Output pulse diameter                | 150 mm                      |
| Focal length                         | Adjustable, 10 m – $\infty$ |
| Receiver telescope primary mirrors:  |                             |
| Number                               | 2                           |
| Viewing directions                   | Vertical and horizontal     |
| Diameter                             | 400 mm                      |
| Receiver telescope secondary mirror: |                             |
| Number                               | 1, switchable               |

The distance  $R_0$  at which, at a given pulse energy, sufficiently high power densities are reached and filamentation starts is controlled by four parameters: initial laser diameter, beam divergence, pulse duration, and chirp. The geometrical parameters are determined by the transmitter telescope, the temporal parameters which lead to “temporal focusing” by the grating compressor. A particular aspect of temporal focusing is the use of an initial chirp. Together with the group velocity dispersion (GVD) of air it allows us to obtain the shortest pulse duration (and thus the onset of filamentation) at the desired location  $R_0$ . For this purpose the compressor is aligned in such a way that a negatively chirped pulse is launched into the atmosphere, i.e., the blue component of the broad laser spectrum precedes its red component. In the near infrared, air is normally dispersive, the red components of the laser spectrum propagate faster than the blue ones. Therefore, while propagating, the pulse shortens temporally and its intensity increases. At the preselected altitude  $R_0$ , the filamentation process starts and white light is generated.

Chirp-based control of the generation of the white-light supercontinuum has been demonstrated using the high-resolution imaging mode of the 2-m-diameter telescope of the Thüringer Landessternwarte Tautenburg (Germany). For these experiments, the Teramobile laser was placed next to the astronomical telescope of the observatory. The laser beam was launched into the atmosphere and the backscattered light was imaged through the telescope. Figure 14.14a shows a typical image at the fundamental wavelength of the laser pulse ( $\lambda = 800$  nm), over an altitude range from 3 to 20 km. In this picture, strong scattering is observed from a cirrus layer at an altitude of 9 km and from a thinner cloud layer around 4 km. In some cases, a scattered signal could be detected from distances up to 20 km. Turning the same observation to the blue-green band (385 to 485 nm), i.e., observing the white-light supercontinuum, leads to the images shown in Fig. 14.14b and 14.14c. As mentioned, filamentation and white-light generation strongly depend on the initial chirp of the laser pulse, i.e., white-light signals can only be observed for adequate GVD precompensation (Fig. 14.14b). With optimal chirp parameters, the white-light channel could be imaged over more than 9 km. It should also be pointed out that, as presented above, the angular distribution of the emitted white light from filaments is strongly peaked in the backward direction, and that most of the light is not collected in this imaging configuration.

Under some initial laser parameter settings, conical emission due to leakage out of the plasma channel could also be imaged on a haze layer,

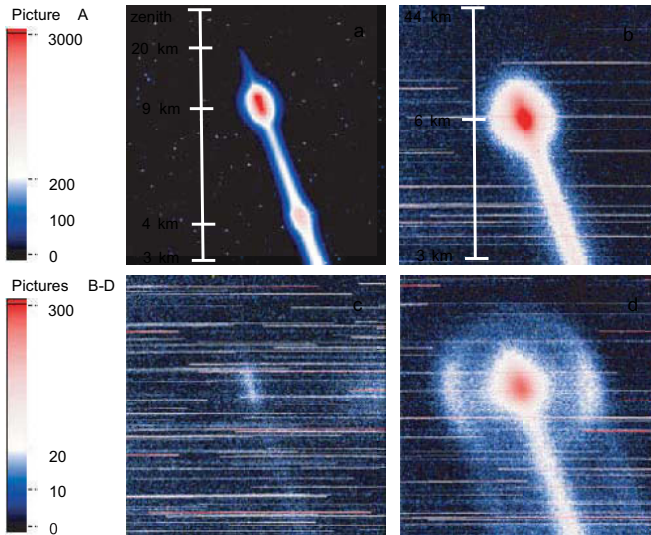


**Fig. 14.14.** Long-distance filamentation and control of non-linear optical processes in the atmosphere [50]. Pictures of the fs-laser beam propagating vertically, imaged by the CCD camera of the 2-m telescope at the Thüringer Landessternwarte Tautenburg. (a) Fundamental wavelength, visible up to 25 km through two layers of clouds. (b) to (d) supercontinuum (measured in the 390–490 nm band) generated by two 600-fs pulses of the same initial peak power, with, respectively, negative (GVD precompensating) (b) and positive chirp (c). The stripes across the images are due to star motion during the acquisition time of several minutes. These results show that white-light generation requires precompensation of the group velocity dispersion (GVD). In picture (d) conical emission appears as a ring on the high-altitude haze layer.

as shown in Fig. 14.14d. Since conical emission is emitted sideways over the whole channel length, the visible rings indicate that under these experimental conditions, the channel was restricted to a shorter length at low altitude.

This fs white-light laser is an ideal source for lidar applications. Linear processes like Rayleigh, fluorescence or Mie scattering return only a small fraction of the emitted light back to the observer. This necessarily leads to an unfavorable  $1/R^2$ -dependency of the received light, where  $R$  is the distance from the scatterer to the observer. When spectrally dispersed, this usually leaves too small signals on the receiver, as arc-lamp-based lidar experiments have shown in the past [57]. Unlike these linear processes, the more pronounced backward emission from white-light channels, as described above (Subsection 14.3.2), allows high spectral resolution of the observed signals, even from long distances. As a

result, it should be possible to obtain spectral fingerprints of atmospheric absorbers along the light path.

Such a white-light-channel based lidar experiment is schematically depicted in Fig. 14.15. It shows the fs laser pulse which, after passing the chirp-generating compressor set, is transmitted vertically into the atmosphere. The backscattered portion of the white light generated in the atmosphere is then collected and spectrally resolved in the lidar receiver.

Figure 14.16 shows examples of spectrally filtered white light lidar returns in three different spectral regions, visible at 600 nm and UV around 300 and 270 nm, averaged over 1000 shots. These profiles of

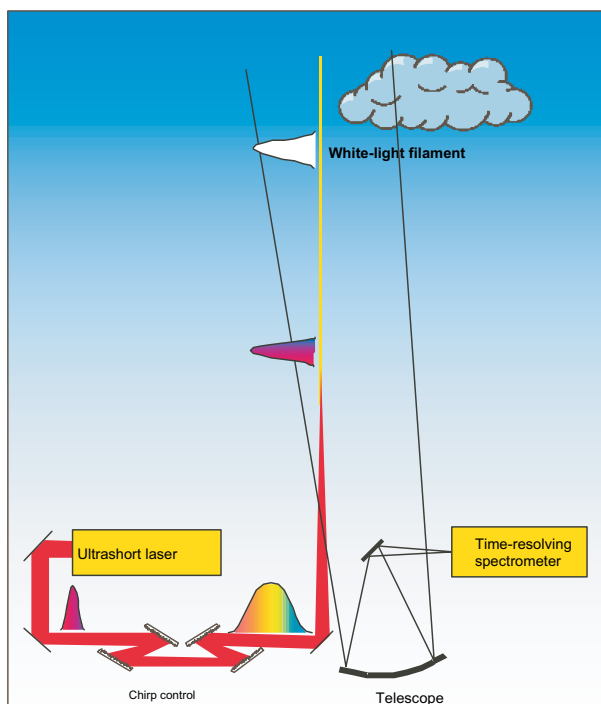


**Fig. 14.15.** Schematics of the white-light TW-laser based lidar experiments [50]. The ultrashort laser pulses are negatively chirped (see text) before launch into the atmosphere. The group velocity dispersion (GVD) in air then compresses the pulse leading to filamentation and supercontinuum generation at a given altitude. The propagation of the white-light laser-pulse is measured range-resolved with the lidar setup. Spectrally resolved lidar measurements of the white-light lidar returns provide simultaneous multicomponent analysis. The location at which filamentation starts can be chosen by the setting of the GVD-compensating chirp.

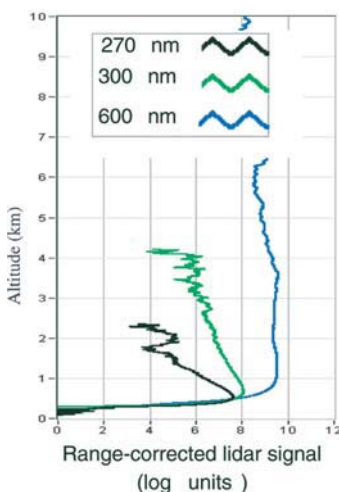


**Fig. 14.16.** Typical white-light lidar returns, at three selected wavelengths (600, 300, 270 nm), showing the stronger atmospheric extinction in the UV due to Rayleigh scattering and ozone absorption (at 270 nm).

white light, remotely generated *in situ*, reveal scattering features of the planetary boundary layer. The faster decrease of the 270-nm signal compared to that at 300 nm is due to the stronger Rayleigh scattering at shorter wavelengths and to stronger absorption by ozone. The white-light spectrum generated over long distances in the atmosphere shows significant differences compared to the spectrum of Fig. 14.11 which was previously recorded in the laboratory [56]. Figure 14.17 displays the white-light spectrum backscattered from an altitude of 4.5 km [60]. The infrared part of the spectrum (taken with filters) is significantly stronger (full line, typically two orders of magnitude higher) than in the laboratory, which is very encouraging for future multi-VOC detection. A *quantitative* explanation of this IR enhancement requires the precise knowledge of the nonlinear propagation of the TW laser pulse which cannot be simulated with sufficient accuracy using the present numerical codes. However, it qualitatively indicates that the pulse shortens and/or splits while propagating, introducing broader frequency components into the spectrum.

On the short-wavelength end of the spectrum (not shown), it was observed that the supercontinuum extends continuously down to 230 nm, the limit being set by the spectrometer. This UV part of the supercontinuum is the result of efficient third-harmonic generation in air [51, 52]

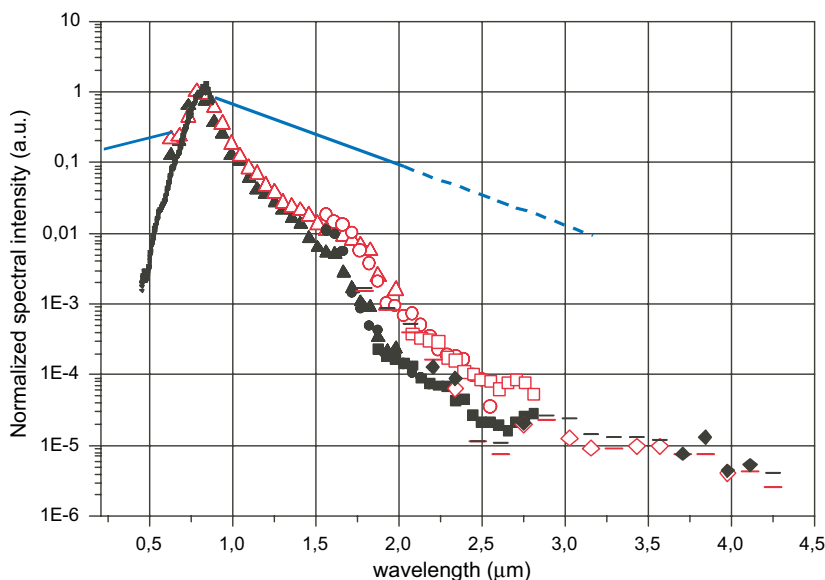


**Fig. 14.17.** Comparison of the supercontinuum spectra measured in the laboratory (as previously shown in Fig. 14.11, red and black) and in the atmosphere at an altitude of 4.5 km (blue) [60]. Long-distance propagation clearly enhances fundamental-to-white-light conversion, especially in the IR (by more than two orders of magnitude at 2  $\mu\text{m}$ ).

and mixing with different components of the VIS-IR part of the spectrum. This opens very attractive applications such as multi-aromatics (benzene, toluene, xylene, ...) DIAL, detection without interference,  $\text{NO}_x$  and  $\text{SO}_2$  multi-wavelength DIAL, and  $\text{O}_3$  measurements in which aerosol interference can easily and automatically be corrected.

Very rich features arise from the backscattered white light when it is recorded across a high-resolution spectrometer, as shown in Fig. 14.18. The spectrum, which was detected from an altitude of 4.5 km with an intensified charge-coupled device (ICCD), shows a wealth of atmospheric absorption lines at high resolution ( $0.01\text{ cm}^{-1}$ ). The excellent signal-to-noise ratio (2000-shot average) demonstrates the advantages of using a high-brightness white-light channel for multi-component lidar detection. The well-known water vapor bands around 720, 830 and 930 nm are observed simultaneously. Depending on the altitude (i.e., the water vapor concentration), stronger or weaker absorption bands can be selected. Figure 14.19 (top) shows a higher-resolution spectrum of the water vapor  $(000) \rightarrow (211)$  transition around 815 nm and Fig. 14.19



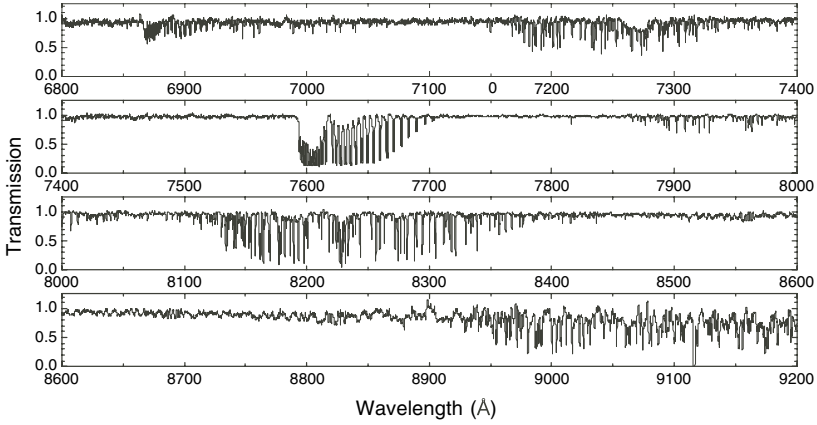


**Fig. 14.18.** High-resolution spectrum of atmospheric absorption measured with the fs white-light lidar from an altitude of 4.5 km [50]. The selected spectral region (660–920 nm) shows the many well-known oxygen and water-vapor bands, all acquired simultaneously.

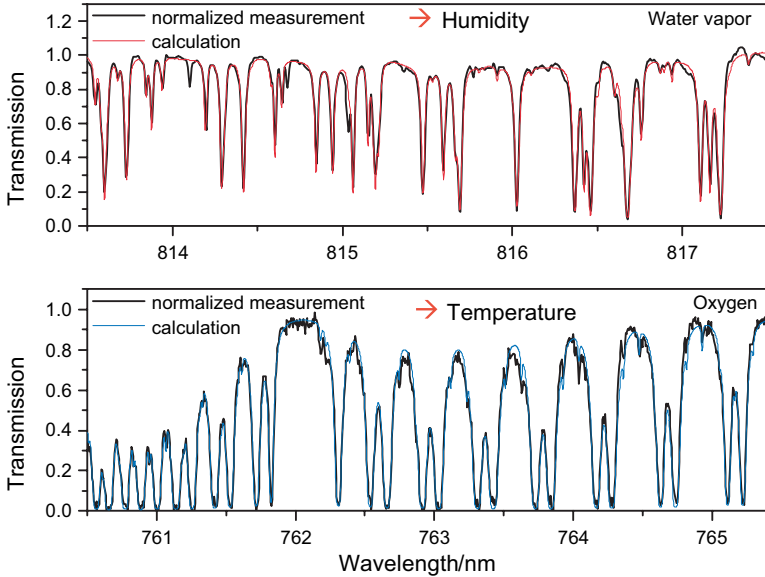


**Fig. 14.19.** Expanded part of the spectrum of Fig. 14.18. A fit using HITRAN data shows that no saturation due to non-linearity occurs. The fit on the many lines allows simultaneous high-precision temperature and humidity lidar measurements.

(bottom) of the  $X \rightarrow A$  transition of molecular oxygen. A fit using the Hitran database is also shown in both cases. It leads to a mean water vapor concentration of 0.4%. Note the excellent agreement with the database, demonstrating that no nonlinear effects or saturation perturb the absorption spectrum. This is explained by the fact that the white light returned to the lidar receiver is not intense enough to induce saturation, and that the volume occupied by the filaments (the white light sources) is very small compared to the investigated volume.

The spectrum used to retrieve the water-vapor concentration contains about 700 data points. The use of so many wavelengths should allow a major improvement in sensitivity as compared to the usual two-wavelength DIALs. A systematic study is in progress to quantify this gain, connected to the obtained signal-to-noise ratio in each spectral element. Using a gated ICCD, the spectrum of the atmosphere can be recorded at different altitudes, yielding range-resolved measurements.

Information about atmospheric temperature (and/or pressure) can be obtained from the observed shapes of the absorption lines. Another possibility is to measure the intensities of the hot bands and to deduce temperature from the ground-state population. As the molecular oxygen spectrum is very well known,  $O_2$  is particularly well suited for this purpose. The access of the whole spectrum should again provide significantly better precision than in former DIAL investigations [61, 62]. The spectrum covered by the white light in Fig. 14.18 gives access to many bands suited to measure the  $H_2O$  concentration and two bands of  $O_2$  ( $(0)'' \rightarrow (0)'$  and  $(0)'' \rightarrow (1)'$  sequences of the  $X \rightarrow A$  transition, around 760 and 690 nm, respectively) to determine atmospheric temperature. The combination of both types of data obtained with good precision could give rise to an efficient future “relative-humidity lidar profiler.”

### 14.3.4 Nonlinear Interactions with Aerosols

Particles are present in the atmosphere as a broad distribution of sizes (from 10 nm to 100  $\mu\text{m}$ ), shapes (spherical, fractal, crystals, aggregates, etc.), and compositions (water, soot, minerals, bioagents such as pollen, bacteria, or viruses, etc.). The lidar technique has shown remarkable capabilities in fast 3-D mapping of aerosols, but mainly *qualitatively* through the measurement of backscatter and extinction coefficients which are hard to relate to other aerosol properties unless

additional information is available about the nature of the aerosols. In order to obtain *quantitative* mappings of aerosols, complementary data from *in situ* instruments such as particle counters, impactors and all kinds of instruments for chemical analysis are often used. For the analysis of lidar data, these data on size distribution and composition must, however, be taken cautiously because of their strong variation in both time and space. Remote range-resolved “all optical” measurement of aerosols without *a priori* assumptions remains therefore an important challenge. Nonlinear spectroscopy with ultrashort laser pulses appears as a promising new technique to fulfill this task, especially for liquid aerosols such as water microdroplets (clouds, fogs) and for other microparticles such as bioagents. In this section, we describe important nonlinear interactions potentially useful for the simultaneous measurement of size and composition of aerosol mixtures and for the identification of one given type of particles within an ensemble.

### Use of the Supercontinuum Produced by SPM in Air

The first approach to using ultra-intense laser pulses in a lidar arrangement for the characterization of aerosol particles is a direct extension of the multiwavelength scattering technique. The extraordinary broadness of the super-continuum, spanning the whole optical range from the UV to the mid-IR, opens several new perspectives. Backscatter and extinction coefficients can now be obtained depth-resolved over the whole continuum, not just for a few individual wavelengths. This is particularly advantageous for mixtures of particles or for unidentified particles for which a multitude of size parameters  $x = 2\pi r/\lambda$  must be addressed. The data inversion could follow the course of sophisticated multiwavelength algorithms already developed for discrete wavelengths (see Chapter 4), which can be much more powerful because of the continuity and greater spectral width of the spectrum. In addition to the experiments made with the Teramobile system [50], some recent lidar measurements of aerosols using the femtosecond supercontinuum generated in rare gas before transmitting were reported [63]. Part of the spectrum might also be analyzed at higher resolution, in an attempt to detect variations in the imaginary part of the refractive index which describes optical absorption, and thus provide additional information on the composition of the particles.

A key parameter in these femtosecond lidar experiments will be the location of the onset and the end of filaments. In particular, if the pulses

are shaped in such a way (negative chirp, see p. 426) that filamentation occurs at short distances and lasts only for a few hundreds of meters, the light scattered back from longer distances can be considered as being scattered linearly. Those data can then be safely inverted with the usual linear lidar algorithms. Conversely, if the laser pulses are initially shaped in such a way that high intensity and filaments are present in the investigated volume, then nonlinear effects occur in the aerosols, and new inversion algorithms must be developed [64, 65]. Examples of these non-linear processes induced in the aerosol particles are presented below.

### **Multiphoton-Excited Fluorescence (MPEF) and Multiphoton Ionization (MPI) in Aerosol Particles**

Femtosecond laser pulses can provide very high pulse intensity at low pulse energy. This allows to induce non-linear processes in particles without deformation by electrostrictive and thermal expansion effects.

The most prominent feature of non-linear processes in aerosol particles, especially in spherical microdroplets, is strong localization of the emitting molecules within the particle and subsequent backward enhancement of the emitted light. This unexpected behavior is extremely attractive for lidar applications. For homogenous spherical microparticles, molecules in certain regions are indeed more strongly excited than others because of the focusing properties of the spherical microresonator. Further localization is achieved by the nonlinear processes which involve the  $n$ th power ( $n \geq 2$ ) of the light intensity  $I$  inside the particle,  $I^n(\mathbf{r})$ ;  $\mathbf{r}$  is the position in the droplet. Because the droplet acts as a spherical lens, reemission from these internal focal points is predominantly in the backward direction. The backward enhancement can be explained by the reciprocity principle [67,68]. Reemission from regions of high  $I(\mathbf{r})$  tends to return toward the illuminating source by essentially retracing the direction of the incident beam that gave rise to the focal point.

We investigated, both theoretically and experimentally, incoherent processes involving  $n = 1$  to 5 photons [66–68]. For  $n = 1, 2, 3$ , we focused on MPEF of fluorophor- or amino acid-containing droplets. For  $n = 5$  (or more) photons we examined laser-induced breakdown (LIB) in water micro-droplets, initiated by multiphoton ionization. The ionization potential of water molecules is  $E_{\text{ion}} = 6.5 \text{ eV}$  [69, 70], so that five photons are required at a laser wavelength of 800 nm to initiate the process of plasma formation. The growth of the plasma is also a

nonlinear function of  $I(\mathbf{r})$ . We showed that both localization and backward enhancement strongly increase with the order  $n$  of the multiphoton process. Both MPEF and LIB have the potential of providing information about aerosol composition.

The strongly anisotropic spontaneous emission of MPEF in a microdroplet was demonstrated on coumarin-510-doped ethanol [67] droplets with radii ranging from 10 to 50  $\mu\text{m}$ . Figure 14.20 shows the angular distribution and the comparison between experimental and theoretical (Lorentz-Mie [66]) results for the one- (400 nm, Fig. 14.20a), two- (800 nm, Fig. 14.20b), and three-photon (1.2  $\mu\text{m}$ , Fig. 14.20c) excitation process. It can be seen that the fluorescence emission is maximum in the direction toward the exciting source. The directionality of the emission increases with the increase of  $n$  because the excitation process involves the  $n$ th power of the intensity,  $I^n(\mathbf{r})$ . The ratio of radiation emitted under  $180^\circ$  and  $90^\circ$ ,  $R_f = U(180^\circ)/U(90^\circ)$ , increases from 1.8 to 9 when  $n$  is changed from 1 to 3. For three-photon excited fluorescence (3PEF), fluorescence from aerosol microparticles is therefore emitted mainly backwards, which is ideal for lidar experiments.

The backward enhancement also depends on the particle relative refractive index  $m$ : the higher  $m$ , the higher  $R_f$ . When excited by one photon at 266 nm,  $R_f$  from dye-doped polystyrene latex (PSL) microspheres ( $m = 1.59$ , typical diameter = 22.1  $\mu\text{m}$ , fluorescence peaked at 375 nm) reaches 3.2 instead of 1.8 for coumarin-510-doped ethanol droplets. Such an enhancement effect is also observed for nonspherical transparent particles like clusters of small (diameter  $< 2 \mu\text{m}$ ) PSL spheres [71]. Another remarkable property is that the backward enhancement is insensitive to the size if the droplet diameter exceeds a few  $\mu\text{m}$ . This was shown by both the calculations for liquid spherical droplets and by experiments on clusters of PSL spheres for which the equivalent diameter was changed from 2 to 10  $\mu\text{m}$  [71]. However, although the  $R_f$  ratio is not sensitive to the particle shape at least for a one-photon excitation process, the high-resolution *2-D angular pattern* in the near-backward direction might be specific of the morphology of the particle.

LIB experiments were made in pure and in saline water droplets. Figure 14.21 shows the white-light ( $500 \pm 35\text{-nm}$  region) angular distribution (in the scattering plane) for an incident intensity of  $1.8 \times 10^{12} \text{ W/cm}^2$ . The observed far-field emission is strongly enhanced in the backward direction and exhibits a narrow secondary lobe near  $150^\circ$ . The agreement between the experimental results and our Lorentz-Mie calculations [68] is excellent. LIB then takes place only at the internal

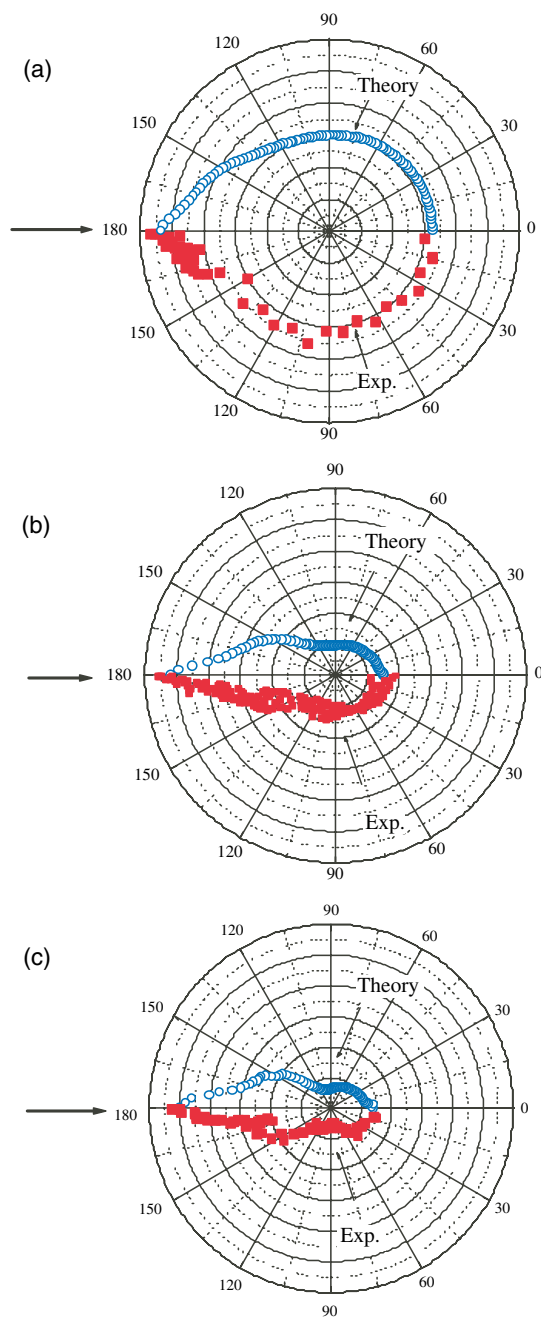


**Fig. 14.20.** Angular dependence of multiphoton-excited fluorescence (MPEF) from microdroplets: (a) 1PEF, (b) 2PEF and (c) 3PEF [66]. A strong enhancement of the emitted light in the backward direction is observed for both 2PEF and 3PEF excitation.

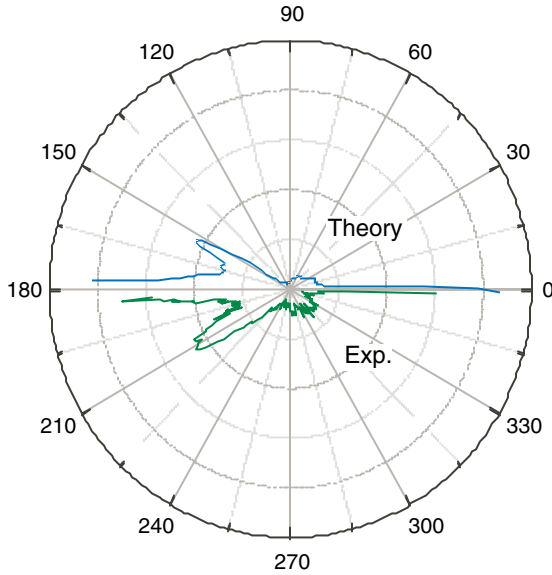


**Fig. 14.21.** White light emitted by laser-induced breakdown (LIB) in water microdroplets [68]. Most of the plasma light is sent back toward the illuminating laser source [backward enhancement  $U(180^\circ)/U(90^\circ) = 35$ ].

hot spot of the droplet, and generates a plasma of nanometric dimensions because of the  $I^5(\mathbf{r})$  dependency of MPI. The white light emitted by the nanoplasma has a ratio  $R_p = U_p(180^\circ)/U_p(90^\circ)$  that exceeds 35 (not shown in the figure because of the chosen scale), i.e., three times higher than for 3PEF.

The spectrum and the related plasma temperature have been measured by using an optical multichannel analyzer (OMA). The broadband visible emission was recorded in the backward direction from pure and saline droplets with various incident intensities. In Fig. 14.22 (curve a), we show that in the case of saline droplets and for an incident intensity  $I_{\text{inc}} = 1.6 \times 10^{12} \text{ W/cm}^2$ , the spectrum can be fitted by a Maxwell–Planck distribution, in agreement with laser-heated plasma emission. Also, when the incident intensity is gradually increased to  $10^{13} \text{ W/cm}^2$  (curves b and c), the emission spectrum shifts toward the blue in a way consistent with an increase of the plasma temperature from 5000 to 7000 K. A similar behavior has been observed for pure water droplets (Fig. 14.22d), but here unexpected and unidentified atomic or molecular lines appear in the spectrum. The shift of the emission maximum is correlated with the change in the angular distribution.

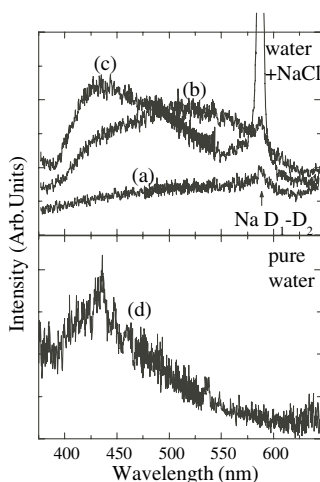


**Fig. 14.22.** Spectrum of the white light emitted by LIB in microdroplets [66]. Upper figure: saline water, displaying the Na lines around 589 nm. Curves (a), (b) and (c) correspond to increasing incoming intensities, which yield increasing temperature Planck radiation curves. Lower figure: pure water.

### Pump-Probe Measurements of Size and Ballistic Trajectories

The very short extension of femtosecond pulses (50 fs correspond to  $11 \mu\text{m}$  in water, i.e., the equator length of a  $1.8\text{-}\mu\text{m}$ -radius sphere) has recently been used to measure the size of microdroplets [72]. Moreover, the trajectories followed by the wavepackets within the microparticle could be clearly identified.

The experimental scheme uses 2PEF (in dye-containing microdroplets) to create an optical correlator between a wavepacket at wavelength  $\lambda_1 = 1200 \text{ nm}$ , and a wavepacket at a different wavelength ( $\lambda_2 = 600 \text{ nm}$ ), each circulating on ballistic orbits. Fluorescence is then recorded as a function of the time delay between the two wavepackets in order to quantify the pathlength traveled inside the particle.

Figure 14.23 (upper curve) displays the fluorescence intensity as a function of the time delay between pulses  $\lambda_1$  and  $\lambda_2$ . The beams were adjusted to hit the droplets ( $r = 700 \mu\text{m}$ , monitored with a CCD camera) tangent to their surface in order to excite surface modes. Positive time delays refer to pulse  $\lambda_1$  reaching the droplet before pulse  $\lambda_2$ . This result, which clearly shows two peaks separated by the round-trip time  $\tau_{\text{RT}}$ , constitutes the first time-resolved observation of the ballistic motion of optical wavepackets within a microparticle. Figure 14.23 (lower curve) displays the same type of scan for a smaller droplet size ( $r = 520 \mu\text{m}$ ),



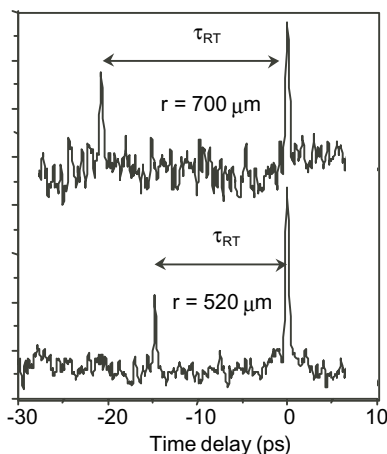


**Fig. 14.23.** Observation of the ballistic motion of femtosecond pulses in microdroplets, using a pump-probe 2PEF technique [72]. The time delay between the two exciting pulses is a signature of the roundtrip time and thus a measurement of the aerosol particle size.

and the consequent decrease in  $\tau_{RT}$ . Since the peak widths of  $\sim 250$  fs do not significantly exceed the correlation time of the two incident pulses, these results demonstrate that femtosecond wavepackets survive evanescent coupling without temporal spreading.

In order to address applications of these pump-probe results to lidar measurements of both size and composition of atmospheric aerosols, time-resolved Lorentz-Mie calculations have been made using plane-wave excitation (70-fs pulses) of a 50- $\mu\text{m}$  droplet [73]. Figure 14.24 shows that similar results are obtained as for the larger droplet excited on the edge, demonstrating the capability of 2PEF pump-probe lidar experiments. Again, the time between two peaks exactly corresponds to the droplet roundtrip time  $\tau_{RT} = 2\pi mr/c$ , where  $m$  is the refractive index). In 2PEF pump-probe lidar experiments, the composition would be addressed by the excitation/fluorescence signatures and the size by the time delay between the two exciting pulses. The high peak contrast in Fig. 14.24 shows that a modulation would still be observable for particles as small as a few micrometers. Shorter pulses could yield measurements of even smaller particles.

### Application to Bioaerosol Detection

Dye-doped microdroplets, because of their high multiphoton absorption cross-sections, are good test cases to demonstrate the advantage of

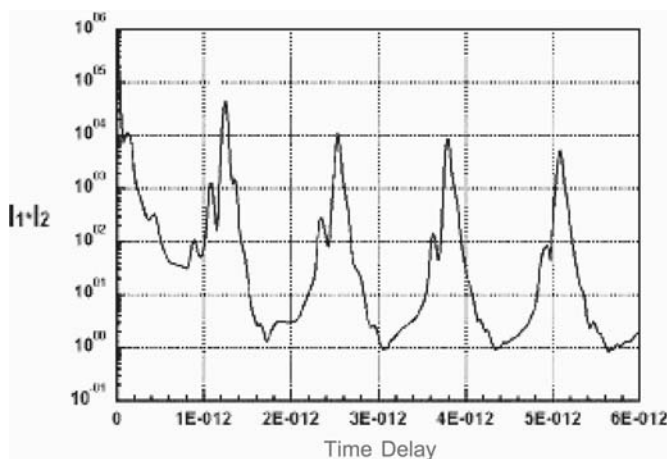


**Fig. 14.24.** Simulations of the pump-probe 2PEF sizing technique for particles of  $50\ \mu\text{m}$  and pulses of 70 fs duration [73]. The time separating two adjacent peaks is representative of the roundtrip time and thus of the particle size. The high contrast obtained appears sufficient to measure particle sizes as small as a few microns. Simultaneous information on the composition is obtained from the fluorescence spectrum.

combining MPEF and lidar techniques to identify the presence of fluorescing aerosols. An attractive application of the combined techniques is bioaerosol detection in the atmosphere. For this purpose, we carried out the first multiphoton excited fluorescence lidar detection of biological aerosols. The particles, consisting of water droplets containing 0.03 g/l riboflavin, a characteristic tracer of bioagents [74, 75] were generated at a distance of 50 m from the Teramobile system. The size distribution peaked around a radius of  $1\ \mu\text{m}$ , a typical size of airborne bacteria. Riboflavin was excited with two photons at 800 nm and emitted a broad fluorescence around 540 nm. This experiment is the first demonstration of the remote detection of bioaerosols using a 2PEF-femtosecond lidar (Fig. 14.25 [50]). The broad fluorescence signature from the particle cloud with typically  $10^4\ \text{particles}/\text{cm}^3$  is clearly seen, with a range resolution of a few meters. As a comparison, droplets of pure water did not exhibit any parasitic fluorescence in this spectral range. However, a background is observed for both types of particles, arising from the scattering of white light generated by the filaments in air. Competition between supercontinuum generation in front of the cloud of particles and 2PEF within the particles appeared critical. A possible solution to this problem is to adapt the initial pulse duration, chirp, and geometrical characteristics of the laser in order that the needed high intensity is

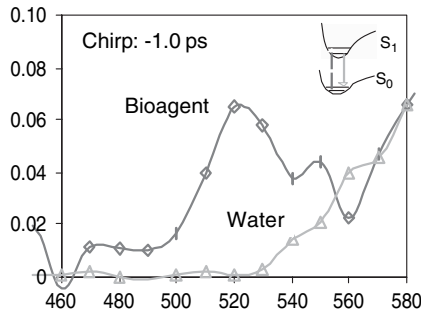


**Fig. 14.25.** Remote detection of biological aerosols using a femtosecond 2PEF lidar technique. An aerosol cloud of riboflavin containing microparticles is detected with the Teramobile at 50 m distance. The 2PEF fluorescence signature allows to identify the composition of the riboflavin particles. In comparison, pure water droplets do not exhibit fluorescence in this region.

reached exactly at the target location. The use of tailored pulses to solve this problem is under investigation; they will also be used to investigate possible simultaneous measurements of size and composition in a pump-probe frame.

MPEF might be advantageous as compared to linear LIF for two reasons: (1) MPEF is enhanced in the backward direction, and (2) the transmission of the atmosphere is much higher for longer wavelengths. For example, if we consider the detection of tryptophan, another typical biotracer that can be excited with three photons of 810 nm, the absorption of the atmosphere is typically  $0.6 \text{ km}^{-1}$  at 270 nm, whereas it is only  $3 \times 10^{-3} \text{ km}^{-1}$  at 810 nm (for a clear atmosphere, depending on the background ozone concentration) [76]. At longer distances this might compensate the lower 3PEF cross-section compared to the 1PEF cross-section. 2PEF could also be an attractive solution if high-power ultrashort lasers around 540 nm could be used. The most attractive feature is, however, the possibility of using pump-probe techniques as described above to measure both size and composition through the length of ballistic trajectories.

## 14.4 Conclusion

In this chapter two new variants of the wide field of lidar have been described, broadband emission lidar with narrowband determination of absorption or BELINDA and ultra-short-pulse, high-power (femtosecond or terawatt) lidar.

BELINDA works with only one laser instead of two. It inherently eliminates problems with the optimum choice of the on-resonance and off-resonance wavelengths and greatly reduces, by more than a factor of ten, the effects of variations in the backscatter coefficients due to changing aerosol concentration gradients. Thus temperature measurements in the planetary boundary layer with high temporal and spatial resolution appear realistic. With a measurement of both moisture and temperature, vertical profiles of the relative humidity can be obtained.

The comparison with DIAL reveals several charming attributes of the BELINDA approach. The future will show whether BELINDA can develop into a technically and commercially viable system able to compete with conventional differential-absorption lidars.

The nonlinear propagation of ultrashort ultra-intense laser pulses provides unique features for lidar applications: a coherent supercontinuum, self-guided, and back-reflected towards the source. Backward enhancement also occurs, for other reasons, for multiphoton-excited fluorescence (MPEF) and laser-induced breakdown (LIB) processes in aerosol particles. These characteristics open new perspectives for lidar measurements in the atmosphere: multi-component detection, reduced spectral interference, better precision through more absorption lines, improved IR-lidar measurements in aerosol-free atmospheres, and remote measurement of aerosol size distribution and composition.

For a widespread application of the technique it is important that our knowledge of the propagation of the laser pulses be further improved, to allow a better prediction of the onset and the length of the filaments and a more precise control of the intensity at each location along the laser path.

Atmospheric applications of filamentation other than lidar are also investigated. The first application concerns lightning control. Filaments are electrically conductive because of the generated plasma and might be used as a laser lightning rod. Promising experiments have been made at a high-voltage facility in Berlin. These experiments have shown that megavolt discharges could be triggered and guided over distances of several meters [77]. Another application concerns triggered nucleation of water droplets. The plasma charges can act as condensation nuclei in saturated atmospheres, in a way similar as in cloud chambers for detecting ionizing particles. Droplet nucleation induced by femtosecond laser filaments was recently demonstrated in the laboratory [50].

As shown above, potential applications of filamentation are numerous and extend over a large number of fields. Ultrashort and ultra-intense

lasers not only provide new perspectives in atmospheric sensing, but open a whole new field in physics and chemistry.

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