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NANOSCIENCE AND TECHNOLOGY

Optical Generation and Control of Quantum Coherence in Semiconductor Nanostructures

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Gabriela Slavcheva
Philippe Roussignol

(Editors)

Optical Generation and Control of Quantum Coherence in Semiconductor Nanostructures

With 137 Figures

 Springer

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To Marta and Metodi Slavchev

Preface

The fundamental concept of quantum coherence plays a central role in quantum physics, cutting across disciplines of quantum optics, atomic and condensed matter physics. Quantum coherence represents a universal property of the quantum systems that applies both to light and matter thereby tying together materials and phenomena. Moreover, the optical coherence can be transferred to the medium through the light-matter interactions. Since the early days of quantum mechanics there has been a desire to control dynamics of quantum systems. The generation and control of quantum coherence in matter by optical means, in particular, represents a viable way to achieve this longstanding goal and semiconductor nanostructures are the most promising candidates for controllable quantum systems. Optical generation and control of coherent light-matter states in semiconductor quantum nanostructures is precisely the scope of the present book.

Recently, there has been a great deal of interest in the subject of quantum coherence. We are currently witnessing parallel growth of activities in different physical systems that are all built around the central concept of manipulation of quantum coherence. The burgeoning activities in solid-state systems, and semiconductors in particular, have been strongly driven by the unprecedented control of coherence that previously has been demonstrated in quantum optics of atoms and molecules, and is now taking advantage of the remarkable advances in semiconductor fabrication technologies.

A recent impetus to exploit the coherent quantum phenomena comes from the emergence of the quantum information paradigm. The scientific effort in this field is focussed on how to exploit the properties of the quantum systems to perform computations. The issues in computation theory are fascinating and recent progress has generated a great deal of excitement. Furthermore, in recent years, a new paradigm focussed on the exploitation of the previously largely ignored spin degree of freedom, has emerged. Spin offers the opportunity to store and manipulate phase coherence over much larger length and time scales than is typically possible in charged-based devices. In this respect, the idea of encoding the quantum information in the spin degree of freedom has been particularly promising and extensively investigated. Furthermore, spin can be accessed through the orbital properties of the electron in

solid state, which in turn can be efficiently manipulated by light according to angular momentum conservation laws, through the optical orientation mechanism. An all-optical implementation of quantum coherent spin control in semiconductor nanostructures, is of particular interest since it takes full advantage of the cutting edge ultrafast laser technologies and enables the implementation of ultrafast schemes for quantum computation.

Although quantum information is undoubtedly a worthy and useful goal in its own right, many more conventional and near-term problems ranging from novel lasers to spintronics are all bound up with issues in coherence. Historically, the quest for the demonstration of fundamental coherent quantum-optical effects in semiconductor systems that have been initially observed in atomic systems has proved to be very successful and has enormous potential for applications. For instance, the idea of achieving Bose-Einstein condensation in solid state at elevated temperatures originates from the cold atom field and was proposed more than 40 years ago. The recent experimental discovery of the room-temperature polariton lasing and the superfluid properties of exciton-polaritons in semiconductor microcavities have opened up new and exciting opportunities for tangible applications of the quantum coherent phenomena of the Bose-Einstein condensation and superfluidity. On the other hand, the exploitation of coherent optical effects, such as electromagnetically-induced transparency and coherent population trapping in semiconductor systems opens up pathways to freeze light in future devices and to build an inversionless laser. This is yet another demonstration of a fruitful transfer of ideas built around the concept of coherence from cold atoms field to the solid state. Although the atomic and semiconductor physical systems are very different, from conceptual and theoretical point of view there are many cognate issues between atomic coherence and the coherence of relatively simple many-body systems such as excitons or exciton-polaritons in semiconductors. The study of these more controllable systems is extremely helpful to interpret and guide work on complex materials with their innumerable confounding issues.

The main focus of this book is the study of the optical manipulation of the coherence in excitonic, polaritonic, and spin systems as model systems for complex coherent semiconductor dynamics, towards the goal of achieving quantum coherence control in the solid-state. The book is intended for graduate students, postdocs and active researchers in the fields of semiconductor quantum optics, nonlinear and coherent ultrafast optical spectroscopies, quantum information processing and quantum computation, semiconductor spintronics, and for physicists and engineers, who want to become familiar with recent experimental and theoretical advances in this frontier research field.

The book provides a selection of review articles written by leading scientists, focusing on various aspects of optically-induced quantum coherence in semiconductor nanostructures. The latest research findings, interpretation and ideas in this rapidly developing field are discussed in four parts: (i) Carrier dynamics in quantum dots; (ii) Optically-induced spin coherence in quantum dots; (iii) Novel systems for coherent spin manipulation, and (iv) Coherent light-matter states in semiconductor microcavities.

Finally, we would like to thank the authors for their enthusiastic response, dedication and full support of the book project until its successful completion. We wish to express our gratitude to R. Murray, E. Clarke, P. Spencer, M. Taylor and E. Harbord for fruitful discussions, providing assistance and valuable feedback on the manuscript. In addition, we are very much indebted to Angela Lahee of Springer-Verlag whose continuous guidance and help during the course of the project were invaluable. Special thanks are due to our families for their encouragement, appreciation and continuous support.

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G. Slavcheva
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Contents

1	Introduction	1
	Gabriela Slavcheva and Philippe Roussignol	
Part I Carrier dynamics in quantum dots		
2	Decoherence of intraband transitions in InAs quantum dots	9
	Thomas Grange, Robson Ferreira and Gérald Bastard	
2.1	Introduction	9
2.2	Electronic states of self-organized quantum dots	10
2.3	Magneto-polaron states in charged QDs	12
2.4	Anharmonic decay of polaron states	14
2.5	Time resolved studies of pure dephasing in QDs	16
2.6	Conclusion	22
	References	23
3	Spectral diffusion dephasing and motional narrowing in single semiconductor quantum dots	25
	Guillaume Cassaboïs	
3.1	Introduction	25
3.2	Theory	26
3.2.1	Random telegraph noise	27
3.2.2	Gaussian stochastic noise	28
3.3	Experiments	29
3.3.1	Unconventional motional narrowing	30
3.3.2	Voltage-controlled conventional motional narrowing	32
3.4	Conclusion	34
	References	35

Part II Optically-induced spin coherence in quantum dots

4	Carrier spin dynamics in self-assembled quantum dots	39
	Edmund Clarke, Edmund Harbord and Ray Murray	
4.1	Introduction	40
4.2	Growth and optical properties of In(Ga)As/GaAs QDs	41
4.3	Spin generation and detection	46
4.4	Spin relaxation and dephasing mechanisms in QDs	48
4.5	Outlook	54
	References	55
5	Optically induced spin rotations in quantum dots	63
	Sophia E. Economou and Thomas L. Reinecke	
5.1	Introduction	63
5.2	Useful concepts	65
	5.2.1 Spin state as vector on Bloch sphere	65
	5.2.2 Composite rotations	65
5.3	rf control of spin in quantum dots	66
5.4	Optical control of spin in quantum dots	67
	5.4.1 Energy levels and selection rules	68
	5.4.2 Optical spin rotations	69
5.5	Optical spin rotation proposals	71
	5.5.1 Optical Stark effect based rotation	71
	5.5.2 Adiabatic approaches to spin rotation	71
	5.5.3 Hyperbolic secant based rotations	72
5.6	Outlook	79
	Appendix	79
	5.6.1 Fidelity	79
	5.6.2 Coherent Population Trapping	80
	References	82
6	Ensemble spin coherence of singly charged InGaAs quantum dots . .	85
	Alex Greilich, Dmitri R. Yakovlev and Manfred Bayer	
6.1	Introduction	85
6.2	Experimental technique	88
6.3	Exciton fine structure	91
	6.3.1 Fine structure of heavy-hole exciton	91
	6.3.2 Linear dichroism in longitudinal magnetic field	93
	6.3.3 Circular dichroism in transverse magnetic field	94
	6.3.4 Spectral dependence of the electron g -factor	95
	6.3.5 Anisotropy of electron g -factor in quantum dot plane	96
6.4	Generation of spin coherence	97
6.5	Mode-locking of spin coherence	100
	6.5.1 Spin coherence time of an individual electron	101
	6.5.2 Mechanism of spin synchronization	102
	6.5.3 Tailoring of ensemble spin precession	104

6.5.4	Temperature dependence of electron spin coherence time	108
6.6	Nuclei induced frequency focusing	109
6.7	Collective single-mode precession	115
6.8	Ultrafast optical spin rotation	119
6.9	Conclusions	125
	References	125

Part III Novel systems for coherent spin manipulation

7	Optically controlled spin dynamics in a magnetically doped quantum dot	131
	Doris E. Reiter, Tilmann Kuhn and Vollrath M. Axt	
7.1	Introduction	132
7.2	Model System of a single dot doped with a single Mn atom	133
7.3	Spin flip in the heavy hole exciton system using π and 2π pulses	136
7.4	Switching into all Mn spin states	140
7.4.1	Switching into spin eigenstates	140
7.4.2	Measurement by pump probe spectroscopy	143
7.4.3	Switching into superposition states	145
7.5	Magnetic field in Voigt configuration	146
7.6	Conclusions	149
	References	149
8	Coherent magneto-optical activity in a single carbon nanotube	151
	Gabriela Slavcheva and Philippe Roussignol	
8.1	Introduction	152
8.2	Problem Formulation	154
8.2.1	Dielectric response function of an isolated SWCNT	157
8.2.2	Optical dipole matrix element for circularly polarised light	158
8.3	Theoretical framework for the natural optical activity	159
8.4	Simulation results for the natural optical activity	163
8.5	Magneto-optical activity of a chiral SWCNT in an axial magnetic field	171
8.5.1	Theoretical model of the nonlinear Faraday rotation in an axial magnetic field	172
8.5.2	Simulation results for Faraday rotation	176
8.6	Conclusions	178
	References	179
9	Exciton and spin coherence in quantum dot lattices	181
	Michal Grochol, Eric M. Kessler, and Carlo Piermarocchi	
9.1	Introduction	182
9.2	Theory	183
9.2.1	Neutral quantum dot lattice	183
9.2.2	Charged quantum dot lattice	191

9.2.3	Neutral quantum dots in lattice of optical cavities	194
9.3	Results and discussion	199
9.3.1	Neutral quantum dot lattice	199
9.3.2	Charged quantum dot lattice	203
9.3.3	Neutral quantum dots in lattice of cavities	205
9.4	Conclusions	210
	References	210

Part IV Coherent light-matter states in semiconductor microcavities

10	Quantum optics with interacting polaritons	215
	Stefano Portolan and Salvatore Savasta	
10.1	Introduction	216
10.2	Electronic excitation in semiconductor	217
10.3	Linear and nonlinear dynamics	222
10.4	Entangled photon pairs from the optical decay of biexcitons	229
10.5	The picture of interacting polaritons	231
10.6	Noise and environment: Quantum Langevin approach	234
10.7	Quantum complementarity of cavity polaritons	243
10.8	Coherent Trapping	248
10.9	Spin-entangled cavity polaritons	250
10.10	Emergence of entanglement out of a noisy environment: The case of microcavity polaritons	251
	10.10.1 Coherent and incoherent polariton dynamics	252
	10.10.2 Results	256
10.11	Outlook	260
	References	260
11	Spontaneous coherence within a gas of exciton-polaritons in Telluride microcavities	265
	Maxime Richard, Michiel Wouters and Le Si Dang	
11.1	Introduction	266
11.2	Formation and steady state of a polariton gas	267
11.3	Momentum distribution, polariton thermalization	269
11.4	Similarities and differences between polariton condensation, polariton lasing and conventional photon lasing	270
11.5	Spatial properties	272
	11.5.1 Gross-Pitaevskii equation	272
	11.5.2 Small size condensate	274
	11.5.3 Polariton condensate in disordered environment	275
11.6	Vortices in polariton condensates	278
	11.6.1 Quantized vortices	279
	11.6.2 Half-quantized vortices	282
11.7	Correlations within a degenerate polaritons gas	283
	11.7.1 Spatial first order correlations	283
	11.7.2 Number fluctuations in a polariton condensate	285

11.8	Conclusion and Outlook	287
	References	288
12	Keldysh Green's function approach to coherence in a non-equilibrium steady state: connecting Bose-Einstein condensation and lasing	293
	Jonathan Keeling, Marzena H. Szymańska and Peter B. Littlewood	
12.1	Introduction	293
12.2	Polariton system Hamiltonian, and coupling to baths	297
12.3	Modelling the non-equilibrium system	298
	12.3.1 Non-equilibrium diagram approach	299
	12.3.2 Mean-field condition for coherent state	300
12.4	Effects of baths on system correlation functions	301
	12.4.1 Decay bath and $\langle \Psi_p \rangle$	301
	12.4.2 Pumping bath and $G_{a^\dagger b}^K$	303
12.5	Mean-field theory and its limits	305
	12.5.1 Equilibrium limit of Mean-field theory	306
	12.5.2 High temperature limit of Mean-field theory - simple laser	308
	12.5.3 General properties of mean-field theory away from extremes	310
	12.5.4 Low density limit: recovering complex Gross-Pitaevskii equation	311
12.6	Fluctuations, and instability of the normal state	312
	12.6.1 Photon Green's functions in the non-equilibrium model . .	312
	12.6.2 Normal-state Green's functions and instability	316
	12.6.3 Normal-state instability for a simple laser	320
12.7	Fluctuations of the condensed system	322
	12.7.1 Finite-size effects – lineshape of trapped system	324
12.8	Summary	326
	References	327
	Index	331

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Chapter 1

Introduction

Gabriela Slavcheva and Philippe Roussignol

The concept of quantum coherence applied to light-matter interactions is central to the present book. It unifies a whole spectrum of coherent optical phenomena observed in different quantum solid-state systems. Quantum coherence is naturally invoked in the description of the quantum dynamics, or the time evolution of quantum systems, under an external perturbation. It has long been recognised that an intimate relationship exists between quantum coherence and quantum correlations and the latter can be used in a rigorous definition of quantum coherence. Depending on the number of coherence conditions satisfied by a succession of correlation functions describing the system, the degree of coherence ranges from various orders of incomplete coherence to full coherence, when the successive correlators satisfy an infinite number of coherence conditions. Dynamical processes in quantum systems are wave phenomena, subject to constructive and destructive interferences and therefore the ultimate goal to achieve control of quantum dynamics requires active manipulation of constructive and destructive interferences.

In the past decade, significant progress has been made in the understanding of quantum correlations, entanglement and decoherence processes in semiconductor nanostructures, towards the ultimate goal of realisation of quantum state manipulation in a controlled fashion. In an attempt to physically achieve active manipulation of quantum dynamics, a new class of quantum coherent control techniques has emerged. Quantum coherent control represents predictable manipulation of the quantum system properties on a time scale shorter than typical material decoherence times, so that the system cannot be affected by its environment. These techniques have initially been applied to atoms and molecules, and most recently to solid state systems. Coherent control in semiconductor nanostructures has attracted significant interest because of the possibility of coherent manipulation of the carrier wave functions on a time scale shorter than typical dephasing times, thus enabling ultrafast optical switching and quantum information processing. Until very recently, the laser tools needed to actively manipulate quantum states were not available. These include the ability to tailor the time and frequency domain structure of optical pulses, to precisely meet the characteristics of the system being manipulated.

Recent progress in ultrafast optical pulse reshaping techniques, for instance, can prove to be of key importance for the realisation of the quantum computation goals.

Amongst the many challenges to be overcome towards the physical realisation of quantum computation is the development, characterisation, modelling and understanding of different candidate physical systems. The physical solid-state systems currently being explored include single-spin systems: ion traps, atomic impurities in semiconductors (e.g. spin in diamond), magnetic dopants, excitons and charged excitons in quantum dots (QDs), and carbon-based materials, such as carbon nanotubes and graphene.

A thorough understanding of the decoherence mechanisms in quantum dots is indispensable for successful physical implementation of any quantum coherence control scheme. A QD is a real solid-state system, and contrarily to atoms is coupled to external reservoirs (e.g. lattice phonons, neighbouring trapped carriers etc.). The first part of the book is dedicated to discussion of the specifics of the carrier dynamics and dephasing mechanisms in quantum dots. The authors of the second chapter, Thomas Grange, Robson Ferreira and Gérald Bastard, focus on the inter-sublevel transitions in doped self-assembled quantum dots as a closer analogue of the atomic transitions, as opposed to the interband transitions for which electron-electron scattering efficiently destroys the coherence. The chapter focusses on the pure dephasing induced by the acoustic and LO-phonon coupling. The latter leads to polaron formation and intraband optical decay through polaron states. In fact, the mechanism of energy relaxation in QDs differs from the one in quantum wells and bulk semiconductors, proceeding via anharmonicity-triggered disintegration of polarons.

QD coupling to its electrostatic environment is a main obstacle for applications such as indistinguishable photon generation and qubit coherent manipulation. The chapter by Guillaume Cassabois tackles, both theoretically and experimentally, the extrinsic dephasing mechanism of spectral diffusion that dominates the decoherence in semiconductor QDs at low temperatures or low excitation power. It also shows how the use of external experimental parameters such as a dc-gate voltage may lead to protection of the zero-dimensional electronic states from outside coupling.

The second part of the book is dedicated to the optically-induced spin coherence in QDs. In fact for obvious reasons the spin degree of freedom is rather well protected and semiconductor spin-based approaches to quantum computation have proved to be particularly promising: currently single spins can be isolated, initialised, coherently manipulated and read out using both electrical and optical techniques. Considerable progress has been made towards full control of the quantum states of single and coupled spins in a variety of semiconductors and semiconductor nanostructures and towards the understanding of the underlying mechanisms of loss of spin coherence in these systems. The global research effort at present is focused on generation and manipulation of multi-spin entangled states, in view of implementation of quantum gate operations in scalable architectures.

Chapter 4 by Edmund Clarke, Edmund Harbord and Ray Murray represents a comprehensive review of the state-of-the-art of the carrier spin dynamics in self-assembled quantum dots, with reference to the physical implementation of a qubit.

In order to accurately model QDs and design appropriate qubit structures, a detailed understanding of the growth and resulting structure and composition of self-assembled QDs, is required. The authors investigate the effect of tailoring the growth conditions, and thereby engineering QD properties, on the spin of confined carriers. The operating conditions that would be required for a QD-based spin qubit in order to maintain the spin coherence, and to allow a sequence of successful quantum gate operations to be performed, are examined in detail. Schemes for spin initialization manipulation and detection are discussed either in single QDs or coupled QD structures and analyzed in the quantum computation context, showing that quantum information applications require high level of spin control. This control can be achieved through the application of time-dependent electric, magnetic fields and lasers and is discussed further in chapter 5, where Sophia Economou and Thomas Reinecke review the theory of the optical methods of spin control in charged QDs, employing quantum optics formalism and with particular emphasis on composite spin rotations. Special attention is paid to the exploitation of a specific class of optical pulses with hyperbolic secant shape, which as the authors show, are promising candidates for coherent spin manipulation. The developed theoretical framework is discussed with reference to relevant experimental results, presented in Chapter 6 by Alex Greulich, Dmitri R. Yakovlev and Manfred Bayer. The collective coherent precession dynamics of the individual electron spins in an inhomogeneously broadened ensemble of singly-charged quantum dots in an external transverse magnetic field is studied by time-resolved pump-probe Faraday rotation. Despite observing the collective dynamics of the whole ensemble, due to synchronisation of certain precession frequencies with the periodic mode-locked laser, it is possible to extract the single spin coherence time. Moreover, by using two synchronized lasers and varying the detuning, the spin polarisation of the quantum dot ensemble can be driven to any point on the Bloch sphere on a picosecond time scale. This represents a significant step forward towards the physical implementation of elementary quantum gate operations on an ensemble of QDs.

Part III of the book discusses newly-emerging materials and systems for coherent optical spin manipulation. These include magnetic dopants in QDs, carbon nanotubes and graphene, and superstructures, such as arrays of nanostructures, e.g. quantum dot arrays or arrays of micro- or nanocavities. Magnetic dopants in QDs are advantageous systems for quantum information applications since the spin of a magnetic impurity can be oriented in an external magnetic field. Chapter 7 by Doris Reiter, Tilmann Kuhn and Vollrath Axt presents a theoretical study of the optically-induced spin dynamics in a single quantum dot, doped with a single Mn atom, in an external magnetic field both in Faraday and Voigt configuration. By using a pulse sequence that manipulates both heavy and light-hole excitons, the proposed model predicts coherent (picosecond) switching of the Mn spin from a given initial state to all other spin eigenstates. Possible spin detection schemes by pump-probe spectroscopy are discussed. One of the major requirements for physical realisation of quantum information processing is the preparation of a coherent superposition of states. In this chapter, the authors show that a proper modification of the switching protocol enables the preparation of a coherent superposition of spin states, with a

magnetic field in Faraday configuration acting as a control parameter. Moreover, in a magnetic field applied in Voigt configuration, the resulting ultrafast Mn spin precession can be controlled by the application of additional optical pulses.

Recent advances in the nonlinear and coherent optics of carbon-based materials, and more specifically carbon nanotubes, with their unique chiral molecular structure, have demonstrated their potential for a wide range of device applications. Exploitation of the coherent optical phenomena in these quantum nanostructures enables development of a novel class of ultrafast polarisation-sensitive optoelectronic devices, e.g. ultrafast optical switches, polarisers etc., based on single carbon nanotubes as basic functional components of an integrated future optoelectronic device. Understanding the origin and the dynamics of the natural optical activity and Faraday rotation in chiral macromolecules in the highly nonlinear coherent regime is therefore of great importance. In chapter 8, written by Gabriela Slavcheva and Philippe Roussignol, a theoretical and computational model for the description of magneto-chiral coherent optical effects in an individual chiral single-walled carbon nanotube is proposed. The way the chirality affects the ultrafast nonlinear optical response of a single nanotube, and the magnitude of this effect in terms of the ellipticity and angle of the optical rotation, at zero-magnetic field and in an external magnetic field are investigated. The model of the coherent carrier dynamics, based on a discrete-level representation of the optically-active states near the band edge, predicts giant natural and magneto-chiral gyrotropy exceeding the one of the superior artificial chiral photonic metamaterials. It also provides a framework for investigation of the time evolution of the optically-induced spin coherence in carbon nanotubes.

A very different approach is adopted in chapter 9 since it deals with the exciton and spin coherence in artificial superstructures with confined photons and excitons having spatial periodicity comparable to their wavelength. Atomic optical lattices have recently been proposed as quantum simulators and used to investigate many-body Hamiltonians. Ordered arrays of nanostructures represent special interest in view of the global scientific effort to manipulate multi-spin Hamiltonians and thus realise spin-entangled quantum gates. The specific systems studied are two-dimensional lattices of neutral and charged quantum dots, embedded in a planar optical cavity or two-dimensional arrays of optical cavities containing each one neutral dot. The chapter provides a natural transition between the topics discussed up to now, predominantly dealing with coherence in individual QDs or ensembles of QDs, and the laser-dot interaction of ordered arrays of QDs embedded in an optical cavity, employing the microcavity polariton modes description. Michal Grochol, Eric Kessler and Carlo Piermarocchi theoretically investigate the optical properties of such periodic superstructures in the strong-coupling regime in the polariton picture. For the case of a lattice of neutral dots in a planar microcavity, the authors show that Bragg polariton modes exist by tuning the exciton and the cavity modes into resonance at high-symmetry points of the Brillouin zone. These localized modes are characterized by an extremely small polariton effective mass, making them the lightest exciton-like particles in solids, and therefore potential front runners for polariton Bose-Einstein condensation at high critical temperature. The array of singly-charged

quantum dots embedded in a planar cavity represents an analogue of a spin lattice which can be used as a high-fidelity conditional phase shift gate. The planar lattice of single-mode cavities, containing one neutral QD each, is used as a model system for investigation of the exciton-photon quantum phase transitions. By controlling the exciton and photon hopping energies, a great variety of coupled fermionic-bosonic quantum phase transitions is demonstrated with reference to interpretation of the polariton BEC.

Part IV of the book is dedicated to coherent light-matter states in semiconductor microcavities with embedded quantum heterostructures. In Chapter 10 Stefano Portolan and Salvatore Savasta adopt an original approach to investigate quantum optical phenomena, typically observed in atomic systems, applying it to exciton polaritons in a microcavity. Control of quantum dynamics of many-particle entangled quantum states requires better understanding of the validity range of the quantum behaviour. In this respect, atom-cavity systems, representing an analogue of the solid-state microcavity systems, have been used to investigate quantum dynamical processes for open quantum systems in the strong coupling regime and to explore quantum behaviour that has no classical counterparts. As a consequence of these efforts, a new field of the semiconductor cavity quantum electrodynamics has recently emerged. The chapter theoretically tackles the challenging problem of manipulation, creation and detection of non-classical states in semiconductor microcavities, employing the general formalism of quantum optics and at the same time properly accounting for the physics of interacting electrons, including the effects of noise and dephasing induced by the electron-phonon interaction and the other environment channels. By including higher-order particle correlations, the authors extend the usual semiclassical description of the linear and nonlinear dynamics of the semiconductor electrons interacting with light, to allow description of quantum optical effects.

Due to their photon component, exciton-polaritons are extremely light particles, which makes them advantageous to the other bosons, since the onset of the quantum degenerate regime (Bose-Einstein condensation (BEC)) in a polariton gas may occur at temperatures as high as room temperature. The emergence of a spontaneous coherence within a gas of exciton-polaritons in telluride microcavities is discussed in Chapter 11 by Maxime Richard, Michiel Wouters and Le Si Dang. In spite of the short lifetime of its constituents, a polariton gas can exist in a macroscopic quantum degenerate coherent state, or can exhibit coherence through polariton lasing, depending on the experimental conditions. The chapter reviews experimental and theoretical works carried out in the last decade, aiming to provide a detailed description of polariton coherent states. The role of finite lifetime is discussed in reference with the inherently non-equilibrium character of a BEC in a driven-dissipative system with comparable timescale of losses and pumping dynamics, as opposed to the equilibrium Bose gas. Furthermore, the authors discuss current state-of-the-art experiments and the theory of the most recently discovered superfluid properties of polariton condensates, namely fully quantized and half-quantized vortices, representing singularities in the phase of the order parameter where the the polariton

density is vanishing. Possible driving mechanisms of formation of vortices in polariton condensates are discussed.

The last chapter of this book can be considered as a theoretical counterpart of the preceding one. The scope of this chapter, written by Jonathan Keeling, Marzena H. Szymańska and Peter B. Littlewood, is the application of the Keldysh Green's function approach to the treatment of coherence in a non-equilibrium steady state with the aim to elucidate the connection and distinction between exciton polariton Bose-Einstein condensation and lasing. Within the adopted theoretical approach, the quantum condensate driven out of equilibrium is considered as an open quantum system coupled to baths, which can transfer energy as well as particles to and from the system. This approach allows us to investigate how the decoherence processes affect the condensation under non-equilibrium conditions. In the limit of high-temperature baths, the behaviour of the simple laser is recovered. This chapter provides an illustration of the application of the non-equilibrium diagrammatic formalism to the treatment of a steady state non-equilibrium system which develops spontaneous coherence. The approach is particularly useful and instructive in highlighting the way the non-equilibrium condensate relates both to equilibrium condensates and to lasers, and to understand the ingredients that make it differ from these limits.

Finally, we hope that the topics discussed in the book will represent interest to the scientific community and the book itself will be a useful asset to the researchers in this rapidly-developing field.

Part I
Carrier dynamics in quantum dots

Chapter 2

Decoherence of intraband transitions in InAs quantum dots

Thomas Grange, Robson Ferreira and G  rald Bastard

Abstract The origin of the dephasing of the S-P intersublevel transitions in semiconductor quantum dots is theoretically investigated. The coherence time of this transition is shown to be lifetime-limited at low temperature, while at higher temperature pure dephasing induced by the coupling to acoustic phonons dominates the coherence decay. Population relaxation is triggered by the combined effects of electron-LO-phonon strong coupling, leading to the polaron formation, and phonon anharmonicity. A good agreement is found between the modelling and temperature dependence of the four wave mixing signal measured in recent experiments.

2.1 Introduction

Because of the discrete nature of the low-lying eigenstates in self-organised quantum dots (QDs), it was believed that these zero-dimensional objects would behave like atoms (the "macro-atom" scheme). Most of the experiments and proposals for using QDs as part of Qubits involve interband transitions where the light promotes

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electrons from the full valence band to the empty conduction band, either through discrete transitions or involving the 2D (wetting layer) or 3D continuum of states. For two recent reviews on QDs, see [1, 2]. These hopes were without accounting for the solid-state aspects of the QDs, namely their coupling to the environment. The electronic degrees of freedom are intimately coupled to the vibration of the atoms and also to the randomly fluctuating electric fields due to the charging and discharging of nearby traps [3](see Chapter 3.). As a result, the macro-atom scheme survives only at very low temperature ($T \approx 10\text{K}$ or so) and the coherence of the ground interband transition decays only within a few ps at room temperature.

A very powerful coherence killer in *interband* excitation of QDs is the carrier-carrier interaction. It is known that the Auger effect is extremely efficient for excited interband transitions [3, 4]. The Auger effect however disappears if one uses intersublevel transitions in QDs doped with one electron (or one hole). Actually, an intraband transition in a one electron doped QD is a real analogue of an atomic transition in monovalent atoms.

We focus here in the intraband transitions in doped QDs. Section 2.2 briefly recalls the main aspects of the bound states of dots hosting one electron. The energetics and intrinsic lifetime (due to anharmonic couplings) of QD polaron states are discussed respectively in sections 2.3 and 2.4. Finally, section 2.5 presents very recent theoretical and experimental results of pure dephasing for intraband optical transitions involving polaron states in self-organised QDs.

2.2 Electronic states of self-organized quantum dots

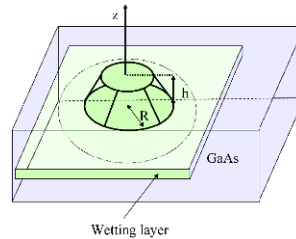


Fig. 2.1 InAs/GaAs quantum dot (schematics)

Let us therefore consider a quantum dot. The Ga(In)As/GaAs self-organised dots are known to be flat objects ($h/R \approx 2\text{-}3\text{ nm}/10\text{nm}$ where h is the dot height and R the basis radius (see Fig. 2.2 for the case of such a dot embedded in a GaAs matrix). It is also known that they are roughly circular. The electronic structure of QDs has been calculated by Stier et al [5, 6] by means of the multi-band envelope function method, while accurate atomistic approaches (pseudo-potential, tight-binding) were undertaken by Williamson and Zunger [7, 8] and by Lee et al [9].

The energy distance between the S -like symmetry envelope ground state in the conduction band and the centre of gravity of the P_x and P_y first excited states is typically ≈ 50 meV for $h \approx 2$ nm and $R \approx 10$ nm [10]. Figure 2.2 shows the results of a numerical computation of the envelope eigenstates of InAs QDs with cylindrical symmetry versus basis radius and maintaining the base angle constant (30°) as well as a fixed height ($h = 2.5$ nm).

One notices below the onset of the 3D GaAs continuum (taken as the energy origin) the existence of a 2D continuum. It is associated with the wetting layer on which the InAs island “floats”. Being only 2.83 thick the wetting layer gives rise only to a weakly bound state for the electron z -motion: the binding energy is very small (≈ 20 meV). True (*i.e.*, along the three directions) bound states, as seen in Fig. 2.2, appear for energies below $E \approx -20$ meV. The actual number of bound states in a QD is a function of its geometry (R and h) and material parameters (confining potential and effective mass). Typically, an InAs-based QD hosts three bound states for electrons in the conduction band: one ground S -like and two P -like states.

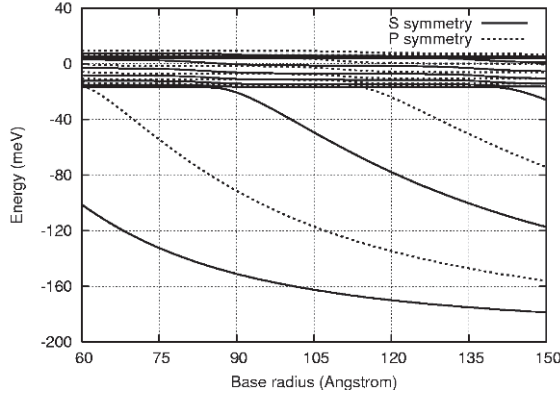
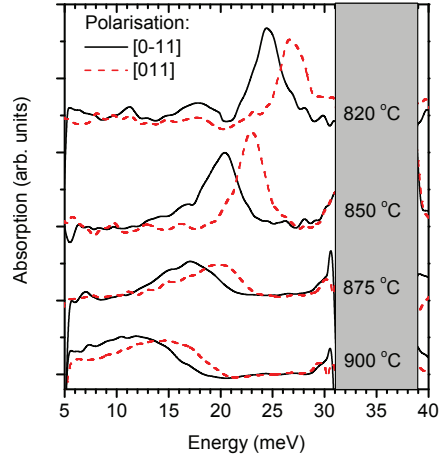


Fig. 2.2 Calculated energy levels in the conduction band of InAs/GaAs QDs versus the base radius. The dot is a truncated cone floating on a one-monolayer thick wetting layer. Solid lines: S levels ; dashed lines: $P_{\pm}$ levels

In Fig. 2.2, the P levels are twice degenerate on account of the in-plane cylindrical symmetry. In fact, there exists a splitting between P_x and P_y (≈ 5 meV) which results from potential energy terms which do not display cylindrical symmetry; for instance the ellipticity of the QD base or/and piezo-electric fields. For modulation-doped with one electron QDs, it is known [11, 12] that there exist two intraband absorptions close in energy. These two absorption lines are respectively allowed when the electric field of the light has a component along the $[1,1,0]$ or the $[1,-1,0]$ crystallographic axis (see Fig. 2.2.).

Fig. 2.3 Absorption of InAs/GaAs QDs multi stacks for different linear polarizations. The spectra correspond to QD samples that have been subjected to post-growth annealing at different temperatures. The S - P absorption lines become broader and red-shifted when the annealing temperature increases.



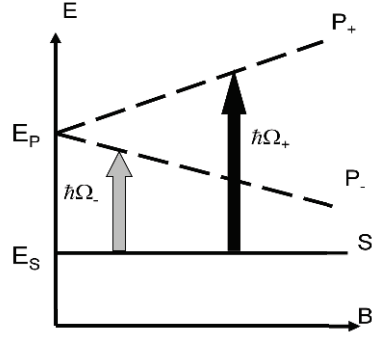
2.3 Magneto-polaron states in charged QDs

The problem of electronic energy levels, relaxation, decoherence in QDs cannot be tackled without a deep analysis of the coupling between confined carriers and phonons. In fact, the energy relaxation problem by phonon emission (the so-called “phonon bottleneck”) [13, 14, 15, 16, 17] remained unsolved until magneto-absorption experiments demonstrated that the weak coupling approach to the electron-phonon interaction in dots, leading to the phonon bottleneck effect, is a model that is essentially inapplicable to the QDs [17]. The polaronic nature of the actual elementary excitation in QDs was evidenced by magneto-optical experiments performed on stacks of InAs QDs.

The magneto-absorption technique probes the optically active eigenstates of the structures. The QDs are loaded with electrons by a controlled modulation (spike) doping of the GaAs barrier. Relaxation effects play a small role (in the magneto-absorption line broadening). The magnetic field (applied along the growth axis in the experiments) acts as a control parameter that fixes the energy distance between the ground state and the first excited electronic state. Because the zero-field size quantization is so pronounced (50 meV or so), the magnetic field acts as a perturbation to the zero-field level scheme. This is the well-known Zeeman effect in the atomic physics except that one deals with flat atoms in the case of InAs/GaAs QDs instead of spherically symmetric atoms. We deal with a orbitally non-degenerate ground state $|S\rangle$ and excited levels $|P\rangle$ which would be twice degenerate at zero-magnetic field in a circular QD (zero-field transitions in Fig. 2.3).

Under the application of a strong magnetic field parallel to the growth direction, the excited states split into $|P_+\rangle$ and $|P_-\rangle$ components (ascending and descending branches in Fig. 2.3) which are separated by the cyclotron energy $\hbar\omega_c$ ($=\hbar eB/m^*$). It is not difficult to account for a slight departure from the circularity. One finds read-

Fig. 2.4 Magnetic field dependence of the lower-lying bound states in a QD. The vertical arrows show the allowed intraband optical transitions for the two counter-rotating circular polarizations. At low temperature, only the ground S -like state is populated.



ily that the ground state is essentially unchanged (apart from a small diamagnetic shift) while the P_+ - P_- degeneracy is lifted even at zero field :

$$\begin{aligned} \epsilon_g(B) &= \epsilon_S + \gamma_S B^2 \\ \epsilon_{\pm}(B) &= \epsilon_P \pm \sqrt{\left(\frac{\delta}{2}\right)^2 + \left(\frac{\hbar\omega_c}{2}\right)^2} + \gamma_P B^2 \end{aligned} \quad (2.1)$$

where δ is the zero field splitting of the P levels and γ_S and γ_P are the (small) diamagnetic contributions to the S and P states respectively. Even at $B = 23\text{T}$ the diamagnetic terms are only a few meVs.

This description of the electronic levels and their fan dispersion under an applied magnetic field corresponds to the so-called "macro-atom" model. Within this model, illumination by far-infrared (FIR) light leads to transitions among different *electronic* levels coupled by the dipolar interaction irrespective of the other degrees of freedom, in particular the vibrations. In particular, intraband magneto-absorption experiments should display two lines at the photon energies $\hbar\omega_{\pm}(B) = \epsilon_{P_{\pm}}(B) - \epsilon_g(B)$ as shown in Fig. 2.3. Figure 2.3 shows the fan chart obtained on a modulation doped QD sample where the QDs contain one electron on average [18]. It is clear that the macro-atom model fails short of explaining these experimental data. In lieu of two branches $\omega_{\pm}$ smoothly varying with B , one finds several anti-crossings superimposed on the two branches $\omega_{\pm}$ of the macro-atom model. A careful analysis of the measurements demonstrates that the anti-crossings occur any time one electronic level intersects the one- or two-longitudinal optical (LO) phonon replicas of another electronic state (*i.e.*, when two macro-atom states differ by $\Delta E = n\hbar\omega_{\text{LO}}$, $n = 1$ or 2 and $\hbar\omega_{\text{LO}} \approx 36$ meV in GaAs and strained InAs). This experimental fact hints that a proper interpretation of the measurements should incorporate the coupling of confined electrons with optical phonons.

In order to interpret the data, a polaron model was built [18, 19] where the Fröhlich interaction $H_{e-\text{ph}}$ is diagonalized between a truncated basis of factorized electron-phonon states. The latter comprises zero phonon level and LO phonon replica of the former. The LO phonons were assumed dispersionless. As a result of the diagonalization of the electron-phonon interaction, the solid lines in Fig. 2.3

were obtained. The experimental results bring evidence that over a broad energy range, typically from 15 meV to about 80 meV, H_{e-ph} mixes very efficiently the factorized states which differ in their electronic quantum number and in their LO phonon occupation number. In other words, it appears that in QDs the discrete nature of the electronic spectrum favors the creation of fairly stable entanglements of electronic and LO phonon states, the *polaron* states. FIR absorption experiments probe such mixed levels, instead of the purely electronic ones. This striking feature is just opposite to the findings in bulk and quantum well structures, where the electron-phonon interaction weakly couples the factorized states and, in fact, can be re-incorporated as a finite lifetime of the factorized states.

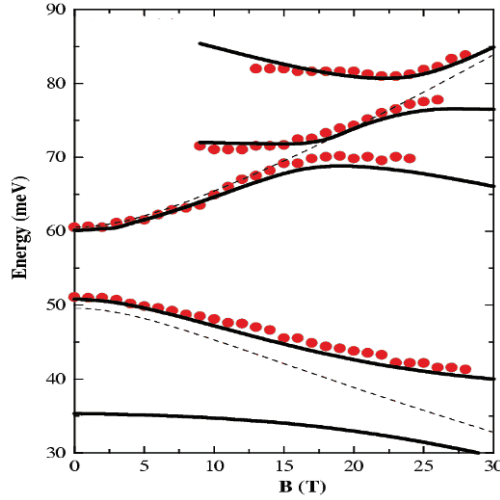


Fig. 2.5 Fan chart of magneto-optical transitions observed on stacks of InAs/GaAs QDs doped with one electron per dot on average. Dashed lines : macro-atom model. Solid lines : polaron model. Symbols : experiments ($T = 4K$). The reststrahlen band is around 36 meV.

2.4 Anharmonic decay of polaron states

The most important aspect of the polaron state, namely the strong coupling of states with different phonon occupancies, can be described by a simple model where only the one-phonon state $|S, 1_q\rangle$ and zero-phonon states $|P, 0\rangle$ are retained in the basis. It turns out that, because LO phonons are dispersionless, the electron-phonon Hamiltonian selects one particular linear combination of $|S, 1_q\rangle$ states to hybridize with each of the discrete $|P\rangle$ state, leading to a final two-level problem. The two resulting polaron levels are then solutions of the coupled Hamiltonian:

$$\begin{pmatrix} E_P & V_{SP} \\ V_{SP} & E_S + \hbar\omega_{LO} - i\Gamma_{LO}/2 \end{pmatrix} \quad (2.2)$$

where V_{SP} is the Fröhlich matrix element involved, and Γ_{LO} is the broadening of the LO phonon energy due to the anharmonicity effects [20, 21, 22]. These polarons will be referred as S-P polarons in the following (P-like polaron will denote the one with a dominant P electronic component). In fact, it is well known that bulk LO phonons have an intrinsic lifetime of a few ps. QD polarons, which are made out of bulk phonons, then become unstable entities. However, the magnitude of the anti-crossing in the magneto-optical transition $S \rightarrow P$ is much bigger than the level broadening: $2V_{SP} \approx 10 \text{ meV} \gg \Gamma_{LO}$. Thus, the anharmonicity effects do not invalidate the polaron model, but instead contribute to its damping into two phonons according to the “chemical” equation:

$$P\text{-like polaron} \rightarrow 2 \text{ phonons} + 1 \text{ electron (S state)} \quad (2.3)$$

This polaron disintegration scheme was experimentally studied [23] recently by means of pump and probe measurements. In these experiments, a free electron laser “FELBE” in Dresden (Germany) delivers linearly polarised picosecond pulses. An initial pulse excites the single electron of a modulation doped QD from the S to the P_x or P_y state and a second pulse probes the recovery of the absorption signal for this same transition as a function of the time delay between the pump and probe pulses. The results of pump-probe experiments performed at $T = 10 \text{ K}$ for different excitation energy are shown in Fig. 2.6 for annealed QDs where the polaron energies are below the Reststrahlen band (see cw spectra in Fig. 2.2). It is seen that the excited state is depleted very quickly (a few ps) near the Reststrahlen band while the relaxation time of the population in the upper state is three orders of magnitude longer when the S - P polaron energy is 14.5 meV .

The modelling of the population relaxation due to anharmonic coupling leads to a lifetime of the polaron state given by the formula [22, 23]:

$$\frac{\hbar}{\tau} = \frac{4\Delta_{SP}^2}{(\hbar\omega_{LO} + \Delta_{SP})^2} \frac{V_{SP}^2}{(\hbar\omega_{LO} - \Delta_{SP})^2 + V_{SP}^2} \Gamma^{ph}(\Delta_{SP}) \quad (2.4)$$

where Δ_{SP} corresponds to the optically probed transition energy between S-like and P-like polaron states with a dominant zero-phonon component. The second term in the above formula corresponds to the weight of the LO-phonon component in the excited polaron. The first term accounts for interferences between resonant and non-resonant Fröhlich interactions. The function $\Gamma^{ph}(\Delta_{SP})$ represents the broadening of the LO-phonon that one has to include in Eq. 2.2 to account for the anharmonicity effects on the polaron. It takes into account all possible disintegration paths of one LO phonon into two other phonons that occur at the polaron energy difference Δ_{SP} . For each decay channel, the relaxation rate strongly decreases with decreasing Δ_{SP} . Three factors combine to produce this decrease: the density of the two acoustic phonon states decreases steeply at low phonon energy, the amount of the one-phonon component decreases from $1/2$ at resonance $\Delta_{SP} = \hbar\omega_{LO} \pm V_{SP}$ to zero

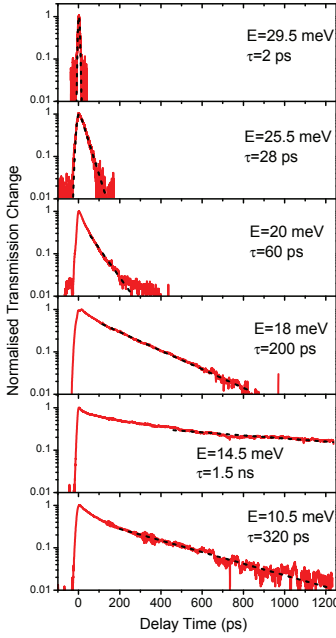


Fig. 2.6 Time dependence of the transient transmission of stack QDs in pump-probe experiments when the S - P energy transition is below the Reststrahlen band.

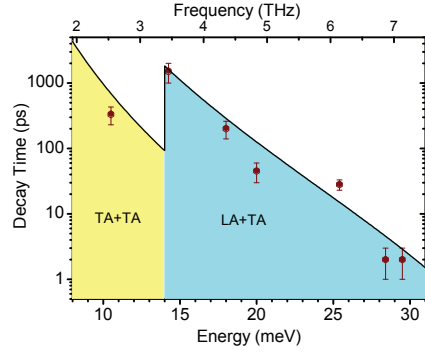


Fig. 2.7 S - P polaron lifetime versus polaron energy below the Reststrahlen band: experiments (symbols) and calculations (solid lines). The dominant anharmonic channels of polaron disintegration are indicated as a function of the polaron energy.

with increasing Δ_{SP} and finally the strength of the anharmonic contributions also decreases with decreasing Δ_{SP} . The important result (see Fig. 2.7.) is the very long population relaxation time of the P -like polaron state. This time is considerably longer than the one found in quantum well-based quantum cascade laser (QCL) devices where the relaxation involves scattering between the 2D in-plane continuum of the lasing levels. This implies that using QDs as the active medium of a QCL may prove to be more efficient in terms of population inversion than using quantum well structures.

2.5 Time resolved studies of pure dephasing in QDs

The S - P polaron coherent polarisation of stacked QDs has been recently studied [24] by using a two-pulse photon echo arrangement in a non-collinear geometry at the free electron laser FELIX in the Netherlands. A ratio 1:2 was used between the

two incoming pulses with wavevectors $\mathbf{k}_1$ and $\mathbf{k}_2$. The intensity of the third order non-linear signal was measured in the $2\mathbf{k}_2 - \mathbf{k}_1$ direction.

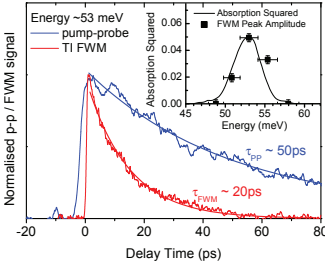


Fig. 2.8 Comparison between the four-wave mixing (FWM) and pump-probe (PP) signals for the same transition energy (53 meV) of the *S-P* transition. Inset: absorption squared and FWM signal intensity of the inhomogeneously broadened *S-P* transition.

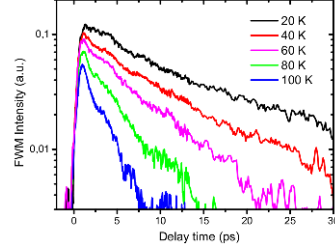


Fig. 2.9 Time dependent four-wave mixing signal (FWM) at different temperatures.

The measurements were performed in the χ^3 regime where the intensity of the four wave mixing (FWM) signal has a cubic dependence on the excitation intensity. A comparison between the FWM and the pump probe signals at the same *S-P* energy of 53 meV and $T = 10\text{K}$ is shown in Fig. 2.8. Both signals decay exponentially with time constants 22 ps and 50 ps, respectively. For an inhomogeneously broadened ensemble of two-level systems, it is known that the coherence lifetime T_2 is equal to four times the decay time of the FWM signal. Hence, at low temperature, $T_2 \approx 90$ ps is nearly twice $T_1 = 50$ ps, the population relaxation time which is the ultimate limit when the pure dephasing processes are negligible. This is no longer the case when the temperature increases, as shown in Fig. 2.9. The decay time of the FWM signal decreases very steeply with increasing temperature (from 22 ps at $T = 10\text{K}$ to 3ps at $T = 100\text{K}$) while the population relaxation time varies much less. This indicates that the pure dephasing processes become dominant when the temperature increases. Such feature is the same as found in the case of the interband transitions [25, 26].

In order to analyze the magnitude and temperature dependence of the coherence time, we have to use the polaron description of the QD eigenstates. However, the intrinsic anharmonicity driven decay of the QD electron and LO-phonon entanglement explains the measured decay only at low temperature. At high temperatures, this decay channel is far too ineffective to be compatible with the observations. Hence, other sources of decoherence have to be invoked to explain the data. Since there is a strong temperature dependence even below 30K, the interaction with the acoustic phonons has to be taken into account. Two contributions show up. (i) Firstly, the acoustic phonon sidebands will contribute to a partial loss of coherence. The phonon sidebands, first analyzed by Huang and Rhys [28], arise from the fact that the equilibrium positions of the atoms are not exactly the same when the electron

is in the excited or ground states, because of the electron - acoustic phonon interaction. In our case we deal with S - P or S polarons but the physics is identical. Since the equilibrium positions are different, there can be electronic optical transitions, triggered by the dipolar coupling to the light, that take place with different phonon occupancies. This is because the phonon wavefunctions (Hermite functions) are not centered at the same origin and, consequently, two phonon wavefunctions that correspond to different phonon occupancies are no longer orthogonal. (ii) In addition, the optically probed S - P_x polaron state is close (a few meVs) in energy from the (S - P_y) polaron state. Therefore, acoustic phonon absorption and emission processes become allowed between these two polaron states. Thus, thermally stimulated escape of a photo-excited carrier from the lower S - P_x to the upper S - P_y polaron state becomes possible and will contribute to the loss of coherence of the S - P_x related transition. In addition, virtual transitions to the other P -like polaron (P_y) state do also have to be considered. These virtual transitions can be considered as a scattering by acoustic phonons thereby inducing a virtual excursion of the polaron to another S - P polaron state through off-diagonal elements of the deformation potential coupling. Virtual transitions do not change the electron populations, but create interactions between the two electronic states involved in the optical transition and the phonon bath, and thus contribute also to the loss of coherence of the optical transition.

In the following, we consider the interaction between the polarons and the longitudinal acoustic phonons via the deformation potential approximation:

$$V = \sum_{i,j} \sum_q M_q^{i,j} (a_q + a_{-q}^\dagger) |i\rangle \langle j| \quad ; \quad M_q^{i,j} = D_c \sqrt{\frac{\hbar q}{3\rho c_s \Omega}} \langle i | e^{iq \cdot r} | j \rangle \quad (2.5)$$

The diagonal components correspond to the Huang-Rhys term and will give rise to the acoustic phonon sidebands while the off-diagonal terms that involve the polarons S - P_x and S - P_y will lead, at the lowest order in V , to the thermal escape of the carrier from the upper state involved in the optical transition. Higher order terms will contribute to the virtual transitions. For the interaction with the acoustic phonons, and far away from the polaron resonance, the electronic components play a more important role than the phonon component (this is exactly the reverse of the population relaxation case where only the one LO phonon part of the polaron wavefunction matters). Hence, from now on we consider only the S , P_x and P_y states. The diagonal components $|S\rangle \langle S|$ and $|P\rangle \langle P|$ involve the same deformation potential D_c since both states belong to the conduction band. This is in striking contrast with the sidebands observed in interband transitions where the initial and final electron states display deformation potentials that have opposite signs. Let us introduce the unperturbed acoustic phonon Hamiltonian:

$$H_{ph} = \sum_q \hbar \omega_q \left(a_q^\dagger a_q + \frac{1}{2} \right) \quad (2.6)$$

We see that the diagonal terms of the electron-phonon interaction can separately be diagonalized when H_{ph} is added. In fact, let us define the new phonon operators:

$$b_q^+ = a_q^+ + \frac{M_{i,q}^*}{\hbar\omega_q} = a_q^+ + S_{i,\bar{q}}^* \quad ; \quad M_{i,q} = M_q^{i,i} = D_c \sqrt{\frac{\hbar q}{3\rho c_s \Omega}} \langle i | e^{iq \cdot r} | i \rangle \quad (2.7)$$

Then, the complete Hamiltonian $H_{ph} + V$ can be exactly diagonalized like:

$$H = H_{ph} + V = \sum_q \hbar\omega_q \left(b_q^+ b_q + \frac{1}{2} \right) - \sum_q \frac{|M_{i,q}|^2}{\hbar\omega_q} \quad \text{with } i = S, P \quad (2.8)$$

As expected the phonons have kept the same vibration frequencies (displacing an harmonic oscillator does not affect its eigenfrequency). But the phonon wavefunctions are not the same. The new eigenstates are:

$$|\tilde{n}_q\rangle = \frac{(b_q^+)^n}{\sqrt{n!}} |\tilde{0}_q\rangle \quad ; \quad |\tilde{0}_q\rangle = \exp\left(-\frac{S_{i,q}^2}{2}\right) \exp(-S_{i,q} a_q^+) \quad (2.9)$$

Because of the modification of the phonon states, the intraband optical spectrum related to the S - P transition can now occur with or without conservation of the phonon occupancy. The phonon conservation gives rise to the zero phonon line (ZPL) and the transitions that involve emission or absorption of one or several phonons give rise to the phonon sidebands. Let us denote by ε the detuning with respect to the purely electronic contribution $\varepsilon_P - \varepsilon_S$. Then the absorption lineshape is given by [24]:

$$A(\varepsilon) = Z e^f(\varepsilon) \quad ; \quad e^f(\varepsilon) = \delta(\varepsilon) + f + \frac{f \otimes f}{2} + \dots \quad (2.10)$$

where the exponential function is taken in a convolution sense, $f(\varepsilon)$ is the function:

$$f(\varepsilon) = \sum_q \frac{|M_q^{PxPx} - M_q^{SS}|^2}{\varepsilon^2} [N(|\varepsilon|) + \Theta(\varepsilon)] \delta(|\varepsilon| - \hbar\omega_q) \quad (2.11)$$

and Z is the weight of the zero phonon line:

$$Z = \exp\left(-\int_{-\infty}^{+\infty} d\varepsilon f(\varepsilon)\right) \quad (2.12)$$

In Eq. 2.10, the i^{th} term corresponds to a transition that involves i -phonon processes (absorption and/or emission). It can be checked that $A(\varepsilon)$ is normalized to 1 and that it can also be rewritten as:

$$A(\varepsilon) = \frac{1}{h} \int_{-\infty}^{+\infty} dt e^{-i\frac{\varepsilon t}{h}} \exp\left[\int_{-\infty}^{+\infty} d\varepsilon' f(\varepsilon') \left(e^{-i\frac{\varepsilon' t}{h}} - 1\right)\right] \quad (2.13)$$

which is the Fourier transform of the time evolution of the S - P coherence [29]. A calculation of $A(\varepsilon)$ convolved with a Lorentzian of linewidth Γ_2 (see below) is shown in Fig. 2.5. The shape differs considerably from the one found for interband transitions. Note that the phonon wings have two clear maxima, which are separated by ≈ 3 meV. Hence they are likely to give rise to observable features in the time evolution of the FWM signal if the damping is not too large. The physical origin of this salient property of the coupling to the acoustic phonons in intraband transitions is the fact that the same deformation potential affects the initial and final states, while different deformation potentials control the electron-phonon couplings in the conduction and valence bands.

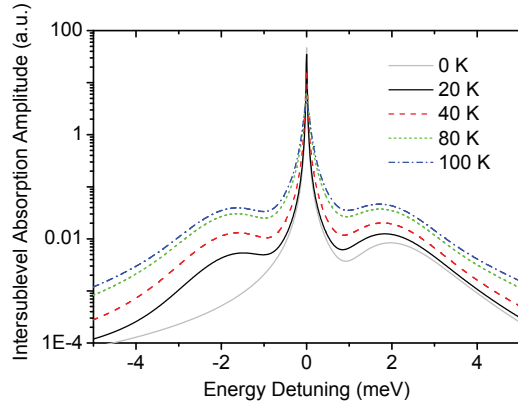


Fig. 2.10 Calculated absorption lineshape of the S - P intersublevel transition versus energy detuning with respect to the zero phonon line (ZPL).

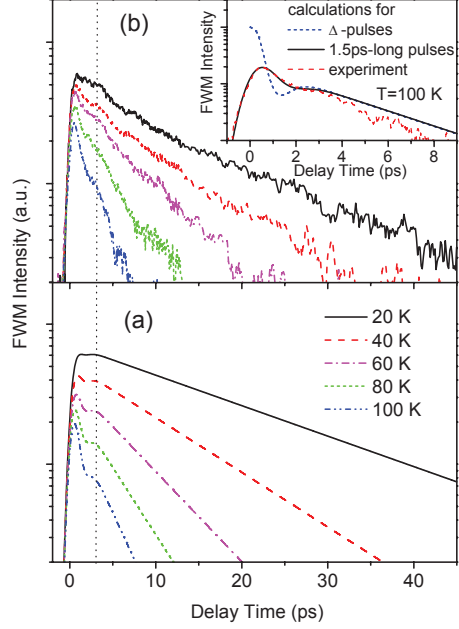
Figure 2.5 shows the time evolution of the FWM signal at different temperatures. We see that the exponential decay is more and more pronounced when T increases. We do see a small feature near 2 ps that we attribute to the peculiar double maxima in the absorption curve versus energy.

The exponential decay of the FWM signal cannot be accounted for only by the diagonal part of the electron-phonon interaction. The ZPL is in this approximation unbroadened and therefore there is only a partial loss of coherence due to the diagonal coupling. In order to obtain further decoherence, we need to take into account thermal escape from the excited state of the transition (the thermal escape from the ground state is negligible for S - P transition energies in the 50 meV range). In fact there are two P levels that are as close as a few meVs. After a delta excitation pulse, the FWM signal is given by the expression:

$$I(t) \propto \Theta(t) \exp \left[-2\Gamma_2 t - 16 \int_{-\infty}^{+\infty} d\varepsilon f(\varepsilon) \sin^2 \left(\frac{\varepsilon t}{2\hbar} \right) \right] \quad (2.14)$$

The broadening Γ_2 can be decomposed in $\Gamma_2 = \hbar/\tau + \Gamma_2^*$, where τ is the anharmonic polaron decay time to the ground state (Eq. 2.4), and Γ_2^* has been calculated by taking into account the off-diagonal part of the electron-phonon interaction up

Fig. 2.11 Temperature dependent FWM signal. (a) Simulations. (b) Experiments. Inset: Calculated FWM signal at 100K for a delta pulse and for a 1.5ps duration pulse compared to the experimental data.



to two-phonon processes [24, 27]. The one-phonon processes correspond to the absorption of an acoustic phonon from the lower P level to the upper one while the two-phonon processes are virtual transitions between the two P levels. We found:

$$\Gamma_2^* = \frac{1}{2\pi} \int_0^\infty d\varepsilon \frac{4\Delta_{PP}^2}{(\varepsilon + \Delta_{PP})^2} \times \frac{\Gamma_{PP}^2(\varepsilon) N(\varepsilon) [N(\varepsilon) + 1]}{(\varepsilon + \Delta_{PP})^2 + \left(\frac{\Gamma_{PP}(\varepsilon) [N(\varepsilon) + 1]}{2} \right)^2} \quad (2.15)$$

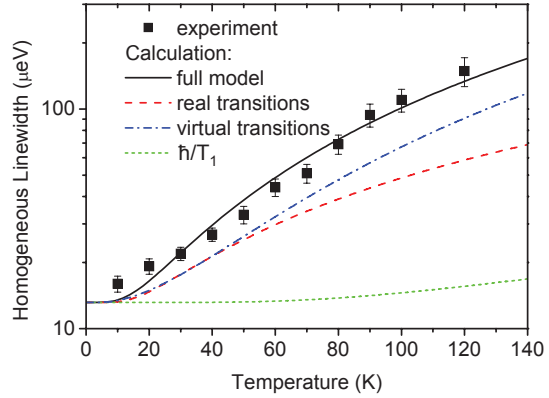
where

$$\Gamma_{PP}(\varepsilon) = \frac{2\pi}{\hbar} \sum_q \left| M_q^{P_x P_y} \right|^2 \delta(\varepsilon - \hbar\omega_q) \quad (2.16)$$

and $N(\varepsilon)$ is the Bose distribution function. It is the pole at Δ_{PP} that corresponds to the absorption of one acoustic phonon while phonons that are not resonant with Δ_{PP} give rise to the virtual transitions between the two P levels. One can show that the phonons with energies between 2 and 3 meV contribute the more to these virtual transitions and their contribution to the dephasing is proportional to $N(\varepsilon)[N(\varepsilon) + 1]$. Therefore, the real or virtual transitions between the two P levels depend strongly on temperature. This is illustrated in Fig. 2.5 where the homogeneous linewidth is plotted versus T . One sees that the contributions of the virtual and real transitions to the dephasing are comparable up to 60K. However, above 60K the virtual transitions are prevalent. The measured and calculated linewidths are in good agreement. Γ_2

increases rapidly from 15 meV at 10 K to 150 meV when $T = 120$ K. This is in marked contrast with the population relaxation time (dotted line in Fig. 2.5).

Fig. 2.12 Temperature dependence of the *S-P* transition linewidth: experiments (closed squares) and calculations including real and virtual transitions between the *P* states. The dotted line accounts for the population relaxation (time T_1) from the upper to the lower state of the transition.



2.6 Conclusion

Time resolved pump and probe and degenerate four-wave mixings experiments have been performed on ensemble of InAs/GaAs self-organized QDs. Experiments clearly show that the energy relaxation in these zero-dimensional objects is of different nature from the one found in bulk and quantum well materials. The formation of polarons between electrons and LO phonons invalidates the usual energy relaxation scheme, namely the irreversible emission of LO phonons by the excited electrons. In QDs the energy relaxation proceeds via the anharmonicity-triggered disintegration of the polarons. The polaron model has proven to be in quantitative agreement with experiments when the QD size increases and makes the *S-P* transition energy to become smaller than the longitudinal optical phonon energy. Very long (ns) relaxation times for electrons in the excited states are found in these broad dots. The coherence of the *S-P* transition energy has been measured and calculated versus temperature. The coherence of intraband transitions behaves in a similar way as found in the case of interband experiment. It is only at very low temperature, say $T < 10$ K, that the pure dephasing mechanisms can be safely neglected.

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Chapter 3

Spectral diffusion dephasing and motional narrowing in single semiconductor quantum dots

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Abstract In this chapter, we address the extrinsic dephasing mechanism of spectral diffusion that dominates the decoherence in semiconductor quantum dots at cryogenic temperature. We discuss the limits of random telegraph and Gaussian stochastic noises, and we describe the general effect of motional narrowing in the context of spectral noise. We emphasize the unconventional phenomenology of motional narrowing in standard semiconductor quantum dots at low incident power and temperature, that makes the quantum dot emission line a sensitive probe of the extrinsic reservoir fluctuation dynamics. We further show that the text book phenomenology of motional narrowing in nuclear magnetic resonance is recovered in quantum dots embedded in field-effect heterostructure. In that case, the electrical control of the mesoscopic environment of the quantum dot leads to conventional motional narrowing where the motion consists in carrier tunneling out of the defects around the quantum dot.

3.1 Introduction

The application of single semiconductor quantum dots (QDs) in quantum information devices is tightly related to the QD decoherence dynamics and to its impact on the device efficiency. In single photon sources, the broadening of the QD emission line beyond the natural linewidth does not restrict the use of these nanostructures [1], whereas it raises severe limitations in the prospect of indistinguishable photon generation [2, 3], or Qubit coherent manipulation

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[4]. One of the most prominent features of QDs, and more generally of solid-state nanostructures investigated for quantum information application, is the strong modification of the decoherence dynamics with the temperature of the surrounding matrix. In semiconductor QDs at room temperature, the decoherence is governed by the intrinsic interaction with acoustic phonons and by the subsequent appearance of lateral phonon side-bands in the optical spectrum. More precisely, the coupling to a continuum of acoustic phonons leads to an emission spectrum that consists in a central zero-phonon line with lateral sidebands, that dominate the optical spectrum at room temperature [5]. On the contrary, at low temperature, the QD matrix contributes to the decoherence dynamics through the extrinsic process of spectral diffusion. Photoluminescence experiments in single QDs have revealed that the QD environment generates fluctuating electric fields which shift the QD line through the quantum confined Stark effect. This so-called spectral diffusion phenomenon was interpreted as due to carriers randomly trapped in defects, impurities in the QD vicinity [6, 7, 8]. This phenomenon with spectral jitters ranging from few tens of μeV to several meV breaks down the simple picture of a two-level system for which the optical line would be given at low temperature by the radiative limit in the μeV range. In this paper, we review the extrinsic dephasing mechanism of spectral diffusion in semiconductor QDs. In Section 3.2, we first present the theoretical framework that allows a comprehensive description of spectral diffusion. In the two limiting cases of random telegraph and Gaussian stochastic noises, we discuss the existence of two fluctuation regimes: the slow modulation one where the QD optical spectrum directly reflects the statistical distribution of the QD emission energy, and the fast modulation one, also called motional narrowing regime, where the QD line is a Lorentzian with a width smaller than the standard deviation of the QD energy. In Section 3.3, we illustrate the rich phenomenology of spectral diffusion dephasing in semiconductor QDs by first showing the experimental evidence for a smooth transition between the two fluctuation regimes. In standard QDs, the motional narrowing regime strikingly occurs in the limit of low temperature or low incident power, in contrast to the text book example of nuclear magnetic resonance. In a second example, we show that motional narrowing can be controlled by a dc-gate voltage for a QD embedded in a field-effect heterostructure. In that case, we observe tunneling-assisted motional narrowing where the motion consists in the tunneling of carriers out of the defects located in the QD mesoscopic environment. The conventional phenomenology of nuclear magnetic resonance is thus recovered and opens the way for a protection of zero-dimensional electronic states from outside coupling.

3.2 Theory

The optical spectrum of a two-level system perturbed by frequency fluctuation was addressed by Kubo in his seminal paper on the stochastic theory of lineshape and relaxation [9]. The central point is the calculation of the so-called relaxation function $\phi(t)$ which corresponds to the Fourier transform of the intensity spectrum, and

which quantifies the accumulated phase because of the random frequency fluctuation $\delta\omega(t)$ around a mean value ω_0 [9]:

$$\phi(t) = \left\langle \exp \left(i \int_0^t \delta\omega(u) du \right) \right\rangle \quad (3.1)$$

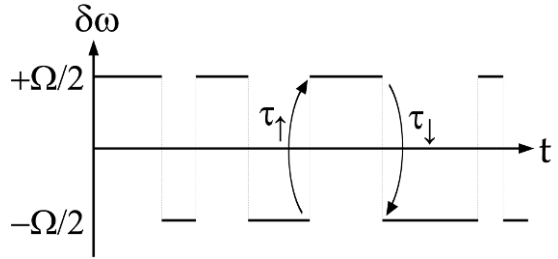
where $\langle \dots \rangle$ denotes the average over different configurations.

Under the assumption of a Markovian modulation, the frequency fluctuation is conveniently analyzed within a model based on a Markov chain composed of an arbitrary number of N independent random telegraphs, as detailed below.

3.2.1 Random telegraph noise

A random telegraph is a two-state jump process that corresponds to a discrete spectral shift $\delta\omega = \pm\Omega/2$ of the optical line. The transition from the upper (lower) state to the lower (upper) one occurs with a probability $dt/\tau_\downarrow$ ($dt/\tau_\uparrow$) in the time interval dt and induces a spectral jump $-\Omega$ ($+\Omega$), as sketched in Fig. 3.1. In the context of spectral diffusion in QDs, the upper (lower) state corresponds to an empty (occupied) defect, $\tau_\downarrow$ ($\tau_\uparrow$) to the capture (escape) time of one carrier in the defect, and Ω to the Stark shift of the QD line due to the electric field created by the charge carrier in the defect located in the QD vicinity.

Fig. 3.1 Frequency fluctuation of the QD emission line as a function of time in the presence of a single random telegraph. Ω is the Stark shift of the QD line due to the electric field created by the charge carrier in the defect located in the QD vicinity. $\tau_\downarrow$ and $\tau_\uparrow$ are the characteristic time constants of the jump processes.



The relaxation function $\phi_1(t)$ of a single random telegraph was calculated by Wódkiewicz *et al.* in the case of symmetric jump processes ($\tau_\downarrow = \tau_\uparrow$ in Fig. 3.1) [10]. In the prospect of a more general theory that would catch the specific physics of semiconductor nanostructures, the former model has been extended to the case of asymmetric two-state jump processes and the expression of the relaxation function $\phi_1(t)$ has been derived when $\tau_\uparrow \neq \tau_\downarrow$ [11]. Following the guidelines of the Kubo theory, the generalized analytical expression of the relaxation function $\phi_1(t)$ reads:

$$\phi_1(t) = G_{+1}(t) - G_{-1}(t) \quad (3.2)$$

with

$$G_\varepsilon(t) = \frac{1 + \varepsilon Y + i\eta X}{2Y} \exp \left[- (1 - \varepsilon Y) \frac{|t|}{2\tau_c} \right] \quad (3.3)$$

where the correlation time τ_c is given by $\frac{1}{\tau_c} = \frac{1}{\tau_\uparrow} + \frac{1}{\tau_\downarrow}$, $\eta = \frac{\tau_\uparrow - \tau_\downarrow}{\tau_\uparrow + \tau_\downarrow}$ characterizes the intensity asymmetry in the emission spectrum, $X = \Omega \tau_c$, and Y is given by $Y^2 = 1 - X^2 + 2i\eta X$ with the condition $\Re(Y) \geq 0$. In the simple case of symmetric jump processes ($\eta=0$), one easily recovers for $\phi_1(t)$ the expression given in Ref [10]. If $\Omega \tau_c \gg 1$, the system is in the so-called slow modulation limit. By taking the Fourier transform of Eq. (3.2) for $\eta=0$, one gets a symmetric doublet structure for the optical spectrum, with two lines split by Ω and a full width at half maximum $1/\tau_c$. In other words, when $\Omega \tau_c \gg 1$, the optical spectrum reflects the statistical distribution corresponding to Fig. 3.1, with a line-broadening given by the fluctuating rate $1/\tau_c$. For asymmetric jump processes, the optical spectrum exhibits an asymmetric doublet where the ratio of the integrated intensity of the two separate lines is given by $\frac{1-\eta}{1+\eta}$. In the fast modulation limit or motional narrowing regime ($\Omega \tau_c \ll 1$), the reduction of the accumulated phase induces the spectral coalescence of the two lines and the doublet structure disappears [9]. The spectrum transforms from a doublet with a splitting Ω to a single Lorentzian line with a width given by the product $\Omega * \frac{\Omega \tau_c}{2}$, thus meaning that the shorter the correlation time, the narrower the line compared to the splitting Ω .

3.2.2 Gaussian stochastic noise

In his pioneering work [9], Kubo analyzed the opposite situation where the frequency fluctuation is characterized by a Gaussian probability density, thus corresponding to a large number of random telegraphs ($N \gg 1$). The derivation of the relaxation function $\phi_{G_N}(t)$ for a Gaussian stochastic noise brought the general theoretical framework for the interpretation of various experiments, and in particular the motional narrowing effect observed in nuclear magnetic resonance [12]. In their pre-Gaussian noise theory, Wódkiewicz *et al.* made the link between random telegraphs and Gaussian stochastic processes by calculating the relaxation function $\phi_N(t)$ for a given number of N identical random telegraphs. Under the assumption of uncorrelated random telegraphs, the relaxation function $\phi_N(t)$ reads $[\phi_1(t)]^N$, and when $N \gg 1$, the relaxation function $\phi_N(t)$ converges to the one obtained for a Gaussian noise $\phi_{G_N}(t)$ [10]:

$$\phi_{G_N}(t) = \exp \left[-\Sigma^2 \tau_c^2 \left(\exp \left(-\frac{|t|}{\tau_c} \right) + \frac{|t|}{\tau_c} - 1 \right) \right] \quad (3.4)$$

which is a consequence of the central limit theorem in the context of spectral noise. The parameter Σ in Eq.(3.4) is the standard deviation of the QD frequency, and in the simple case of symmetric jump processes ($\tau_\uparrow = \tau_\downarrow$), Σ is given by $\frac{\sqrt{N}\Omega}{2}$ [10]. In

order to complete the generalization of the spectral diffusion theory and address the case of Gaussian stochastic processes with asymmetric jump events, the general expression of Σ has been derived when $\tau_{\uparrow} \neq \tau_{\downarrow}$, and the expression of the frequency modulation amplitude Σ reads [13]:

$$\Sigma = \frac{\sqrt{N}\Omega}{\sqrt{\frac{\tau_{\uparrow}}{\tau_{\downarrow}}} + \sqrt{\frac{\tau_{\downarrow}}{\tau_{\uparrow}}}} \quad (3.5)$$

which reaches its maximum value $\Sigma_s = \frac{\sqrt{N}\Omega}{2}$ when $\tau_{\uparrow} = \tau_{\downarrow}$ and vanishes in the limit of strong asymmetric jump processes ($\tau_{\uparrow} \gg \tau_{\downarrow}$ or $\tau_{\uparrow} \ll \tau_{\downarrow}$).

If $\Sigma\tau_c \gg 1$, the system is in the slow modulation limit, in analogy to the single random telegraph phenomenology described above. The relaxation function has a Gaussian decay so that the optical spectrum has a Gaussian profile that reflects the Gaussian distribution law of the spectral noise. The full width at half maximum depends solely on the frequency modulation amplitude and is given by $2\sqrt{2\ln 2}\Sigma$.

In the fast modulation limit $\Sigma\tau_c \ll 1$, the relaxation function dynamics is strongly modified by motional narrowing, and it is characterized by an exponential decay. In the Fourier domain, the optical spectrum corresponds to a Lorentzian line with a width $2\Sigma^2\tau_c$, that is, the shorter the correlation time the narrower the optical spectrum.

In nuclear magnetic resonance, the transition from the slow modulation limit to the fast modulation one was observed on increasing the temperature [12]. This configuration corresponds to the particular case of symmetric jump processes ($\tau_{\uparrow} = \tau_{\downarrow}$) where the frequency modulation amplitude is constant with a value given by $\frac{\sqrt{N}\Omega}{2}$. As a matter of fact, the transition from $\Sigma\tau_c \gg 1$ to $\Sigma\tau_c \ll 1$ implies that τ_c decreases as a function of temperature, which is due to the thermal activation of the nuclei motion in nuclear magnetic resonance [9]. In the case of spectral diffusion in semiconductor QDs, we will see in Section 3.3 that the motional narrowing phenomenology is more complex because the frequency fluctuation dynamics is not restricted to the case of symmetric processes ($\eta=0$).

3.3 Experiments

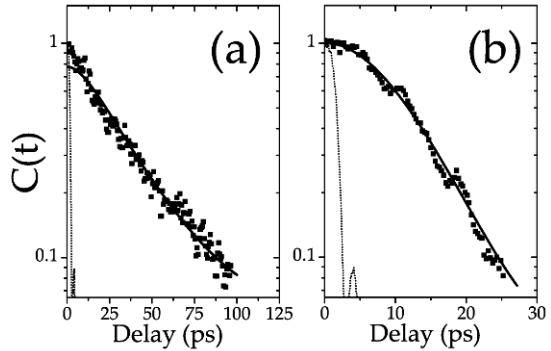
We illustrate the various aspects of motional narrowing in semiconductor QDs by presenting measurements performed in self-assembled InAs/GaAs QDs grown by molecular beam epitaxy in the Stranski-Krastanow mode. The photoluminescence signal arising from a single QD is analyzed by means of Fourier-transform spectroscopy, which allows a high-resolution sampling of the Fourier-transform of the intensity spectrum on typically thousands of points [14]. This value is by two orders of magnitude larger than the average number of illuminated pixels in a charge-coupled device in the case of standard multi-channel detection in the spectral domain. The Fourier-transform technique is implemented in the detection part of the

setup where the photoluminescence signal passes through a Michelson interferometer placed in front of a grating spectrometer. The signal is detected by a low noise Si-based photon counting module. A translation stage varies the time t for propagation in one arm of the interferometer and one records interferograms of the photoluminescence emission $I(t)=I_0(1+C(t)\cos(\omega_0 t))$, where I_0 is the average photoluminescence signal intensity, ω_0 the central detection frequency, and $C(t)$ the interference contrast which corresponds to the modulus of the Fourier transform of the optical spectrum, i.e. with the notation above $C(t)=|\phi_N(t)|$. The implementation of this technique in single QD spectroscopy allows a precise determination of both width and shape of the emission line in order to accurately study the spectral diffusion phenomenon and the related QD decoherence.

3.3.1 Unconventional motional narrowing

We present in this section the experimental evidence for a crossover from the fast to the slow modulation limits. We show that a smooth transition between a Lorentzian line-profile and a Gaussian one is induced on increasing the incident power or the temperature for a QD in a simple heterostructure [13]. The existence of motional narrowing at low incident power or low temperature is a striking manifestation of the asymmetry of the jump processes for spectral diffusion in semiconductor QDs.

Fig. 3.2 Interferogram contrast $C(t)$ of the photoluminescence signal of a single InAs/GaAs quantum dot at 10K, on semi-logarithmic plots, for two different incident powers: 0.18 (a), and 2.88 kW.cm⁻² (b). Data (squares), system response function (dotted line), theoretical fits (solid line) obtained by the convolution of the system response function with $\phi_{G_N}(t)$.

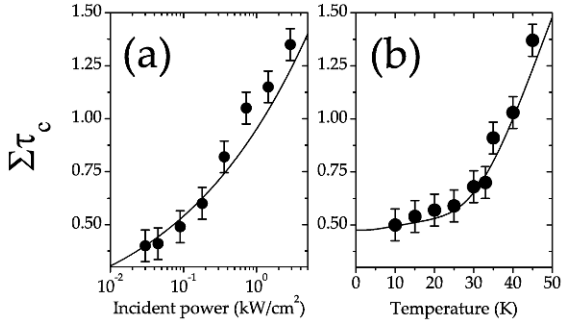


In Fig. 3.2 we display the measured (squares) interference contrast $C(t)$ for the emission spectrum of a single InAs/GaAs quantum dot at 10K, on semi-logarithmic plots, for two different incident powers: 0.18 (a) and 2.88 kW.cm⁻² (b). We first observe that the coherence relaxation dynamics becomes faster for large incident powers. Moreover, there is an important modification in the decay of $C(t)$. At low power (Fig. 3.2(a)), the interference contrast decay is quasi-exponential with a time constant of 29 ps, thus corresponding to a quasi-Lorentzian profile with a full width at half maximum of 45 μ eV. At higher power (Fig. 3.2(b)), the interference contrast

decay is predominantly Gaussian, thus corresponding to a quasi-Gaussian profile, with a full width at half maximum of $155 \mu\text{eV}$.

A quantitative interpretation of the measurements is achieved by comparing the experimental data with the convolution of $\phi_{G_N}(t)$ (Eq. 3.4) with the system response function (dotted lines in Fig. 3.2) which is obtained under white light illumination. The calculated fits are displayed in solid line in Fig. 3.2 and an excellent agreement is reached with increasing values of $\Sigma\tau_c$ of 0.6 in (a), and 1.35 in (b). In Fig. 3.3(a) the whole set of values of $\Sigma\tau_c$ is displayed as a function of the excitation density, on a semi-logarithmic scale. This parameter characterizes the shape of the emission spectrum and the crossover from the exponential to the Gaussian decoherence dynamics occurs around $\Sigma\tau_c \sim 1$. Its gradual increase demonstrates the transition from the fast modulation limit to the slow modulation one when increasing the incident power. Similar results are obtained by means of temperature-dependent measurements where the transition from Lorentzian to Gaussian line-profiles occurs on increasing the temperature, with values of $\Sigma\tau_c$ as a function of the temperature displayed in Fig. 3.3(b). As a matter of fact, it shows that motional narrowing occurs for low excitation and low temperature.

Fig. 3.3 Fitting parameter $\Sigma\tau_c$ versus incident power (a) and temperature (b). Data (symbols), and calculations (solid lines) are plotted as a function of incident power on a semi-logarithmic scale (a), and temperature (b).



The extracted values of $\Sigma\tau_c$ are confronted with calculations performed with Eq. (3.5), and a fair agreement is observed for the parameters $\hbar\Sigma_s \sim 400 \mu\text{eV}$, $\tau_\downarrow \sim 10$ ps, and $1/\tau_\uparrow = (1/\tau_0)\sqrt{P}$ in Fig. 3.3(a) where $\tau_0 \sim 1.6$ ns and P is in unit of $1 \text{ kW}\cdot\text{cm}^{-2}$ [13]. The asymmetry between the power dependences of $\tau_\downarrow$ and $\tau_\uparrow$ stems from the existence of different microscopic processes. The escape rate dependence on incident power is characteristic of Auger-type processes. In the elastic collision of two carriers where one is ejected from the trap while a delocalized carrier relaxes in energy, the escape rate is proportional to the density of delocalized carriers, which increases with incident power. As far as $\tau_\downarrow$ is concerned, the constant value of 10 ps stems from an optical-phonon assisted capture in the traps around the quantum dot [13]. For the temperature-dependent measurements (Fig. 3.3(b)), the escape rate varies with temperature according to $1/\tau_\uparrow = n_1(T)/\tau_1 + n_2(T)/\tau_2$ with $\tau_1 \sim 35$ ns, $\tau_2 \sim 10$ ps, $E_1 \sim 1$ meV, and $E_2 \sim 20$ meV, and where the Bose-Einstein occupation factor $n_i(t)$ given by $1/(\exp(E_i/kT)-1)$ accounts for the absorption of a phonon of

mean energy E_i during the thermally-activated escape of the carriers out of the defects [13].

The asymmetry of the capture and escape mechanisms appears as the fundamental reason why motional narrowing strikingly occurs when decreasing the incident power or the temperature. If both processes had the same efficiency ($\tau_{\uparrow}=\tau_{\downarrow}$), one would have $\tau_c=\tau_{\uparrow}/2$ and $\Sigma=\Sigma_s$, so that the spectral modulation amplitude would not depend on the time constant $\tau_{\uparrow}$. Therefore, the ratio $\Sigma\tau_c$ could only decrease when increasing the reservoir excitation. This situation corresponds to the well-known phenomenology in nuclear magnetic resonance where the activation of the nuclei motion induces the motional narrowing effect. In the present case where $\tau_{\downarrow}/\tau_{\uparrow}\leq 10^{-2}$, we are in the opposite regime where the correlation time is merely constant with relative variation smaller than 10^{-2} whereas the spectral modulation amplitude shows a steep increase with $\tau_{\downarrow}/\tau_{\uparrow}$ ($\Sigma\propto\sqrt{\tau_{\downarrow}/\tau_{\uparrow}}$). The ratio $\Sigma\tau_c$ thus increases when increasing the reservoir excitation with incident power or temperature. As a matter of fact, this non-standard phenomenology explains the observation of Lorentzian profiles in single QD spectroscopy with a width which is not given by the intrinsic radiative limit, but by the extrinsic reservoir fluctuation dynamics [15]. In the prospect of reducing the environment-induced decoherence in quantum information devices, the question then arises whether motional narrowing could be controlled by other parameters than incident power or temperature, in order to recover the conventional phenomenology of nuclear magnetic resonance and achieve an inhibition of dephasing.

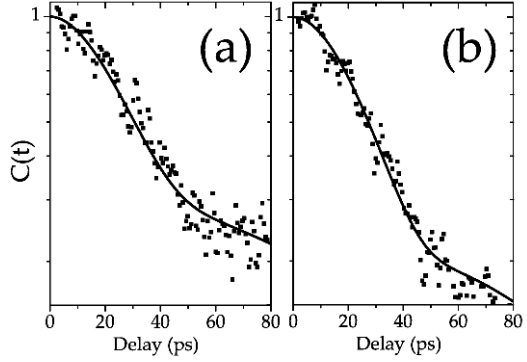
3.3.2 Voltage-controlled conventional motional narrowing

Such a perspective was explored by embedding single QDs in a field-effect heterostructure in order to study the control of the spectral diffusion dynamics with a dc-gate voltage [11].

The sample consists of InGaAs QDs that are separated by a 25 nm GaAs layer from a highly n-doped GaAs substrate. The QDs are capped by 15 nm of GaAs, followed by a 75 nm AlGaAs blocking barrier and finally 60 nm of GaAs. After growth, a 10 nm thick semi-transparent Ti Schottky contact was deposited on the top surface. In order to study single QDs, 400 nm diameter apertures are opened lithographically into a 200 nm thick Au mask [11]. By varying the gate voltage applied between the back and Schottky contacts, one can tune the QD electron states relative to the Fermi energy and thus control the electron occupation in the QDs in order to perform a high resolution study of the neutral exciton line as a function of the gate voltage.

In Fig. 3.4, we display the interferogram contrast $C(t)$ on semi-logarithmic plots, for the neutral exciton line at 30K for two gate voltages of -0.24 (a) and -0.2 V (b). These measurements reveal that the coherence decay at -0.24 V (Fig. 3.4(a)) is slower than the one at -0.2 V (Fig. 3.4(b)), and consequently that the linewidth decreases as a function of the electric field with a total reduction of 20% in the

Fig. 3.4 Interferogram contrast $C(t)$ of the photoluminescence signal of a single InGaAs QD at 30K, on semi-logarithmic plots, for two gate voltages: -0.24 V (a), and -0.2 V (b). Data (squares), theoretical fits (solid line).



whole bias range (Fig. 3.5(a)). These measurements are in strong contrast with the well known tunneling-assisted broadening observed in quantum wells, or in QDs in the regime of photo-current spectroscopy [16, 17]. In fact, the field-induced narrowing observed in Fig. 3.5(a) arises from the electrical control of spectral diffusion, and more precisely from motional narrowing assisted by tunneling. A quantitative interpretation of these experimental data is reached in the framework of the pre-Gaussian noise theory [11]. The complex mesoscopic environment is described by a composite relaxation function where the $\phi_{G_N}(t)$ component (Eq. 3.4) depends on gate voltage through the escape time $\tau_{\uparrow}$. In Fig. 3.4, the calculated contrasts are displayed in solid line and an excellent agreement is obtained thus showing that the model accounts for the variation of the whole emission spectrum (shape and width) with gate voltage. More precisely, the complete set of data as a function of voltage and temperature is fitted by taking $\hbar\sqrt{N}\Omega=56 \mu\text{eV}$, $\tau_{\downarrow}=45 \text{ ps}$, and by only varying $\tau_{\uparrow}$ with the values indicated in Fig. 3.5(b). The constant time $\tau_{\downarrow}$ (shown by a dashed line in Fig. 3.5(b)) is consistent with multi-phonon assisted capture in defects [18].

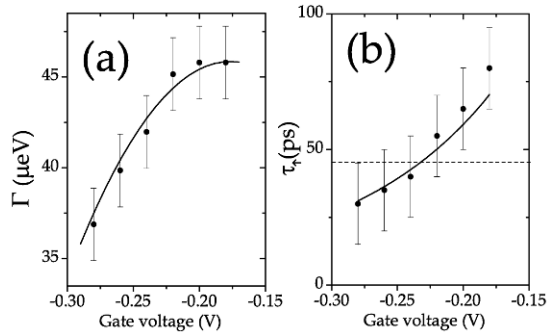


Fig. 3.5 (a) Linewidth versus gate voltage at 30K. Data (symbols), theoretical fits (solid line). (b) Tunneling time $\tau_{\uparrow}$ versus gate voltage. Data (symbols), theoretical fit (solid line). The horizontal line indicates the capture time $\tau_{\downarrow}=45 \text{ ps}$.

In Fig. 3.5(b), a systematic decrease of the escape time $\tau_{\uparrow}$ is observed with the reverse bias, as expected for field-induced tunneling. In analogy to the thermionization of deep centers [19], these variations are interpreted by taking a phonon-

assisted tunneling rate that is proportional to the transmission through a triangular barrier due to a static electric field [19]. The data in Fig. 3.5(b) are reproduced by taking $\tau_{\uparrow} = \tau_{\infty} \mathbb{T}(F)^{-1}$ where $\mathbb{T}(F)$ is the barrier transmission given by $\exp\left(\frac{-4\sqrt{2m^*E_{io}^3}}{3\hbar eF}\right)$ with F the electric field strength along the growth direction, m^* the effective mass and E_i the effective ionization energy. We fit our data with $E_{io}^e=245$ meV in the case of electrons ($m^*=0.07m_0$) or $E_{io}^h=145$ meV for holes ($m^*=0.34m_0$), $\tau_{\infty}=3.5 \times 10^{-5}$ ps. The large values of E_{io} suggest that the spectral diffusers are deep defects, in agreement with the recent experimental evidence by deep level transient spectroscopy of the coexistence of deep levels with optically active InAs QDs [20]. This field-induced narrowing effect demonstrates the achievement of the text book phenomenology of nuclear magnetic resonance where the linewidth decreases by increasing the motion. In our solid-state system, the motion is tunneling controlled by a dc-gate voltage, and it induces an inhibition of dephasing in our single QD-device, that could be implemented in electrically-pumped single photon sources.

3.4 Conclusion

In this chapter, we have addressed the extrinsic dephasing mechanism of spectral diffusion that dominates the QD decoherence at cryogenic temperature. We have discussed the limits of random telegraph and Gaussian stochastic noises, and described the general effect of motional narrowing in the context of spectral noise. We have emphasized the unconventional phenomenology of motional narrowing in standard semiconductor quantum dots at low incident power and temperature, that makes the quantum dot emission line a sensitive probe of the extrinsic reservoir fluctuation dynamics. We have shown that the text book phenomenology of motional narrowing in nuclear magnetic resonance is recovered in QDs embedded in field-effect heterostructure. In that case, the electrical control of the QD mesoscopic environment leads to motional narrowing where the motion consists in carrier tunneling out of the defects around the QD. This effect opens the way for a protection of zero-dimensional electronic states from outside coupling through a control of motional narrowing with external experimental parameters such as a dc-gate voltage.

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Part II
Optically-induced spin coherence in
quantum dots

Chapter 4

Carrier spin dynamics in self-assembled quantum dots

Edmund Clarke, Edmund Harbord and Ray Murray

Abstract Exploitation of spin in the solid state has attracted considerable recent interest for spintronics and quantum computing applications. Semiconductor nanostructures offer improvement in spin relaxation and spin coherence times due to localization of carriers, which leads to the suppression of traditional relaxation and dephasing mechanisms. Quantum dots (QDs), with 3D localization of carriers, are particularly strong candidates for solid state qubits. In this chapter, we investigate how the properties of self-assembled QDs influence the spin of confined carriers, and how these properties can be tailored by choice of appropriate growth conditions. Spin lifetimes of single carriers and excitons are enhanced in QDs but significant spin relaxation and dephasing mechanisms remain, including exchange interaction with other carriers, hyperfine interaction with nuclei and multiphonon processes. These are reviewed and strategies to minimize or eliminate them are discussed. The exploitation of spin coherence in single QDs and coupled QD structures is introduced, with reference to the requirements for implementation of quantum computing schemes.

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4.1 Introduction

One of the main aims of modern physics research is to understand and exploit the quantum mechanical features of a system as well as its classical features. An example of this is in the development of quantum information processing (QIP), where in order to circumvent some of the limitations of classical computation based on digital bits in states 0 or 1, operations are based on quantum bits (qubits) with a superposition of quantum states $|0\rangle$ and $|1\rangle$:

$$|\psi\rangle = \alpha|0\rangle + \beta|1\rangle \quad (4.1)$$

For effective QIP, we need to be able to initialize individual qubits, manipulate them through operations upon single and multiple qubits and read out their final states. We also need the quantum coherence of the qubits to persist for sufficient time for multiple operations to be performed on them [1]. As a figure of merit, a qubit should be able to perform at least 10^3 error-free operations, and ideally towards 10^6 error-free operations in order for a quantum computer to be competitive with its classical counterparts [2]. However, we reach a trade-off between the coherence time of a qubit and its accessibility for initialization and readout. For example, photons are robust against decoherence but can be difficult to address. Although much of the research in these areas uses optical systems [3] or isolated atomic or molecular systems, for example in ion traps [4, 5], where the interaction with the system and its environment can be carefully controlled and minimized, solid state systems are particularly attractive for practical applications because of their compactness, scalability and mature fabrication technologies. However, one of the main challenges when using solid state systems is the interaction of the quantum species with the environment.

Localized electron spins in semiconductors are attractive candidates as possible qubits since they are relatively easy to address optically over short timescales, they can be manipulated using applied electric or magnetic fields or optical pulses and they can exhibit sufficiently long coherence times to be of practical use. The $|0\rangle$ and $|1\rangle$ states of the qubit could be encoded using the spin-up or spin-down states of the electron:

$$|\psi\rangle = \alpha|0\rangle + \beta|1\rangle \equiv \alpha|\uparrow\rangle + \beta|\downarrow\rangle \quad (4.2)$$

The localization of carriers is beneficial for addressing the qubit, and also suppresses the principal spin decoherence mechanisms that occur in a semiconductor. Characteristic timescales for spin relaxation and decoherence are substantially increased compared to those for electron spins in bulk semiconductors, but significant decoherence mechanisms remain, and indeed can be enhanced due to the confinement of the carriers. Three-dimensional carrier localization can be achieved either by the incorporation of impurity atoms [6] or by confinement in semiconductor heterostructures known as quantum dots (QDs). Examples of QDs include those defined by lithography [7] or electrostatically, using surface gate structures to modulate an underlying two-dimensional electron gas (2DEG) [8], colloidal nanocrystals

[9] or monolayer fluctuations in ultrathin quantum wells [10], but in this chapter we focus predominantly on self-assembled QDs formed due to strain interactions during epitaxial crystal growth, concentrating on the In(Ga)As/GaAs material system. These self-assembled QDs have a number of advantages: because of their smaller size compared to those realized by lithography or defined by electrostatic gates, the energy separation between the quantized levels in self-assembled QDs is relatively large, allowing increased operating temperatures. Also, self-assembled QDs may be optically addressed and can be incorporated into device structures during a single growth process.

In this chapter we will examine the operating conditions that would be required for a QD-based qubit in order to maintain the spin coherence to allow a sequence of successful QIP operations to be performed. Spin initialization and detection methods are discussed, and the main spin relaxation and dephasing mechanisms relevant to QDs are examined, focusing on exchange interactions with other carriers in the QD and the hyperfine interaction between carriers and nuclei. Strategies to circumvent these spin loss processes are also suggested. We then briefly consider ways to manipulate the spin in QDs, leading to possible operations on QD-based spin qubits. However, in order to accurately model QDs and design appropriate qubit structures, a detailed understanding of the growth and resulting structure and composition of self-assembled QDs is required, and we introduce this in the next section.

4.2 Growth and optical properties of In(Ga)As/GaAs QDs

Self-assembled QDs are formed by strained layer heteroepitaxy via the Stranski-Krastanov growth mechanism. In this case growth of a material with a mismatch in lattice constant with respect to the substrate initially proceeds by formation of a strained two-dimensional layer known as the wetting layer (WL). The strain builds up with increasing deposition of material and at a critical thickness, θ_{crit} , that is dependent on the lattice constant mismatch, it becomes more energetically favorable for strain to be relieved by the formation of three-dimensional islands. For a limited range of additional coverage beyond θ_{crit} , the islands are dislocation-free [11, 12] and when overgrown (capped) by subsequent material will form nanoscale inclusions within the bulk matrix that can provide three-dimensional quantum confinement of carriers.

For the InAs/GaAs material system there is a 7% lattice mismatch between InAs and GaAs, so that θ_{crit} will be around 1.7 ML (monolayers) for InAs deposition on GaAs at 500°C [13, 14, 15]. The number, size, shape and composition of the islands in the ensemble are a consequence of the interplay between kinetics and thermodynamic effects during growth, and so can be controlled by choice of the growth conditions. As an example, figure 4.1 illustrates the variation in island density with InAs growth rate at a constant growth temperature of 500°C. In this way the QD density can be controllably varied by over two orders of magnitude [16, 17]. Figure 4.1 also shows representative atomic force microscopy (AFM) images of the

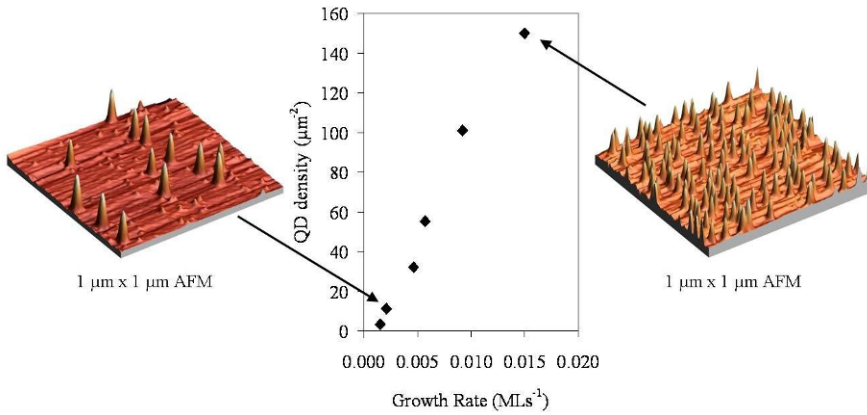
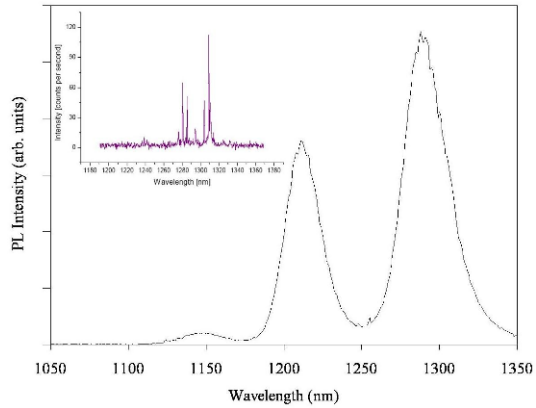


Fig. 4.1 Variation in InAs/GaAs QD density with growth rate at a growth temperature of 500°C, with 1 $\mu\text{m} \times 1 \mu\text{m}$ atomic force microscopy images obtained from samples grown at a growth rate of 0.002 MLs^{-1} (left) and 0.015 MLs^{-1} (right).

InAs/GaAs islands obtained at low growth rate (left) or higher growth rate (right). The highest growth rate shown in figure 4.1 is still around a factor of 10 lower than is often used for QD growth, but this is chosen to provide a high degree of uniformity in island size. There is a variation in the size of the islands, which follows an approximately Gaussian distribution, and the placement of the islands on the surface is apparently random. All of these features have a significant influence on the ability to address single QDs for potential applications but can be optimized by a variety of growth and processing strategies. Capping the islands results in major changes in their size, shape and composition. The island height is rapidly reduced in the early stages of capping, as material comprising the islands diffuses and mixes with the surrounding material [18, 19, 20, 21]. Due to preferential growth of GaAs on the WL, which is still lattice-matched to the underlying substrate, rather than on top of the InAs islands that are partially relaxed towards the bulk InAs lattice constant [22], larger, In-rich islands are less prone to collapse and will retain a higher In content once capped [23]. Intermixing during capping can also be suppressed by reducing the growth temperature, although this may be at the expense of increased defect formation due to poorer GaAs quality [24]. Once capped, InAs/GaAs QDs provide a three-dimensional confinement potential for electrons and holes that will produce discrete electronic states, with interband emission resulting from parity-conserved recombination of excitons [25]. Figure 4.2 shows a low temperature (10 K) photoluminescence (PL) spectrum obtained from an ensemble of InAs/GaAs QDs grown at a very low growth rate (0.002 MLs^{-1}), which yields a low density of QDs (around 13 μm^{-2} , as shown in the AFM image in figure 4.1) with ground state emission at 1300 nm. Because of the low density of QDs, significant excited state emission is observed (first excited state at 1210 nm, second excited state at 1150 nm) due to Pauli blocking of carriers, even at moderate excitation intensities. The emission from the ensemble is inhomogeneously broadened due to the variation in size and

composition of the QDs, giving an approximately Gaussian lineshape for each transition. Inset to figure 4.2 is a micro-PL spectrum obtained from the same sample, once $1\text{ }\mu\text{m}$ -diameter mesas have been etched in order to isolate emission from a few QDs. Now the emission is comprised of sharp lines arising from recombination of individual excitons. The linewidth of the excitonic emission lines from QDs can be extremely sharp for a solid state system, on the order of μeV at low temperatures [26]. Coulombic effects will have a significant influence over the emission energy of excitons, so that emission from charged excitons and multiple exciton complexes will be shifted in energy with respect to the neutral, single exciton (X^0) emission energy by up to a few meV, and the emission from various charged or multiple excitons can be distinguished by variation of the excitation power [27]. Alternatively, the QDs can be placed in a Schottky diode structure, which allows controllable population of the QDs by electrons or holes depending on the applied bias [28, 29, 30]. The influence of applied electric or magnetic fields can lead to resolvable Stark shifts [31, 32] or Zeeman splitting [33] respectively, on the order of meV in emission energy, and can also be used to distinguish different exciton complexes.

Fig. 4.2 10 K PL spectrum obtained from a low density ensemble of InAs/GaAs QDs (InAs growth rate 0.002 MLs^{-1}). Inset: Low temperature (4 K) micro-PL spectrum obtained from a $1\text{ }\mu\text{m}$ diameter mesa etched into the same sample, showing sharp emission lines from single QDs.

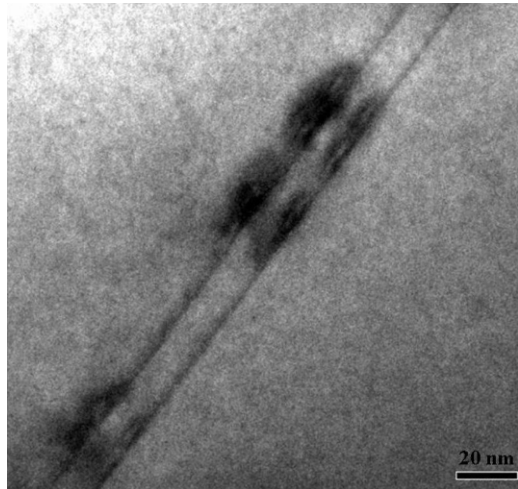


Accurate modeling of the electronic structure of In(Ga)As/GaAs QDs is difficult because of the uncertainties in the size, shape, composition and strain state of the QDs, although multiple band $\mathbf{k}\cdot\mathbf{p}$ or pseudopotential models have been employed with reasonable success [34, 35, 36, 37, 38]. Experimental approaches including high-resolution transmission electron microscopy (TEM) [39, 40, 41], scanning tunneling microscopy (STM) [42, 43] and X-ray diffraction [44] have been used to determine structural information about QDs and show that they are well approximated by lens-shaped or truncated pyramid shapes, with typical heights of 2-10 nm and diameters of 20-40 nm. The composition of QDs can be more difficult to determine (for example the contrast in TEM images is a result of contributions from both composition and strain) but studies indicate a non-uniform InGaAs composition with increasing In content towards the top of the QD [42, 44]. This composition gradient

can arise from diffusion of Ga from the substrate into the QD during growth [45] and also from In segregation [46] and its magnitude is dependent on the In content of the QD [47]. It influences the optical properties of the QD: for a pyramidal QD of uniform InGaAs composition, hole wavefunctions would be expected to be confined towards the base of the QD due to their increased effective mass, with the electron wavefunctions located towards the apex, resulting in a reduced electron-hole wavefunction overlap and a permanent dipole in the QD. Photocurrent measurements demonstrate the existence of this dipole but show that it is in the opposite direction than would be expected for uniform composition [48]. However, eight band $\mathbf{k}\cdot\mathbf{p}$ and pseudopotential calculations taking into account a compositional gradient demonstrate that the hole wavefunction is confined towards the base of the QD and the electron is confined towards its apex and so replicate the observed dipole orientation [49] and can also simulate changes in the emission energy due to changes both in the average In content of a QD and in the In distribution profile [50]. Preferential In migration along the $[110]$ direction during QD growth and particularly during capping can lead to an elongation of QDs in this direction [51, 52]. This leads to a reduction in the symmetry of the confining potential of the QDs and results in a fine structure splitting, δ_{FS} , of exciton emission that is observable by single QD spectroscopy [53], and is typically on the order of tens of μeV [54]. This has implications for the detection of spin in QDs using polarized light and on electron-hole exchange interactions, as discussed later in section 4.4. In order to minimize δ_{FS} , post growth annealing of QDs has been used to alter their size, shape and composition [54, 55, 56, 57], or the asymmetry in the QD potential can be compensated by applying lateral electric [58, 59] or magnetic fields [60] or a uniaxial stress along the $[110]$ direction [61]. In order to achieve the interaction between neighboring qubits that is necessary for a variety of QIP operations, QDs must be placed close enough together for efficient electronic coupling to occur (on the order of 10 nm or less). Current demonstrations of QD-based qubits have only involved one or two interacting QDs, but scalability for a practical quantum computer will require many qubit operations, which would need controlled interactions between many QDs, and this presents a significant technological challenge. As illustrated in the AFM images of self-assembled QDs in figure 4.1, the average separation between QDs is in excess of 50 nm for many growth conditions, so lateral coupling between QDs in the same layer is unlikely. By appropriate choice of growth conditions (using high growth rates, low growth temperature and moderate InAs coverage), the density of self-assembled QDs can be increased up to $1000\ \mu\text{m}^{-2}$ [62] and at these densities the interdot separation is reduced sufficiently for lateral electronic coupling to be expected, but addressing individual QDs would be extremely difficult. Also, from the AFM images in figure 4.1 we can see that the QDs are apparently randomly located on the surface. This makes it difficult to address specific QDs (especially within device structures), which has an impact on device fabrication yield if it is necessary to place single QDs precisely within a structure. Post-growth location of QDs by micro-PL can be employed [63], but numerous groups are also investigating the placement of QDs by growth on patterned substrates (for a review, see [64]). Although significant progress has been made in determining suitable growth con-

ditions for nucleation of QDs at controlled sites on patterned substrates, it can be difficult to achieve similar control over the properties of these QDs (for example the emission energy) as is possible with self-assembled QDs, and their proximity to defects or interfaces introduced by the patterning process can reduce their radiative efficiency. Both single QDs and clusters of QDs may be formed around features on patterned substrates [64, 65, 66], although there has only been limited investigation of in-plane electronic coupling between these QDs [67, 68, 69]. Another approach to achieve lateral electronic coupling between QDs is to grow on vicinal or high index substrates, the surfaces of which will have regular monolayer step-edges. Preferential nucleation of QDs at these step-edges result in the formation of lateral QD chains [70, 71], which would be attractive for some proposed qubit architectures [72].

Fig. 4.3 Bright field TEM image obtained from a cross-section of two closely-stacked InAs/GaAs QD layers. Image provided by S. Kadkhodazadeh and D. W. McComb, Department of Materials, Imperial College London.



Although lateral coupling of QDs can be problematic, self-assembled QD growth is particularly suitable for forming vertically-coupled QD pairs and QD stacks, by exploiting the strain interactions occurring during growth of closely-separated QD layers. For sufficiently small separations between QD layers (typically <30 nm), strain from underlying QDs will provide preferential nucleation sites for QDs in an upper layer, leading to vertical correlation of QDs [73]. This is illustrated by the bright field TEM image of a cross-section of a pair of InAs/GaAs QD layers shown in figure 4.3. By stacking QD layers in this manner, it is possible to very precisely control the separation between QDs. However, growth of stacked layers is considerably more complex than growth of single layers and it is exceedingly difficult to obtain nominally identical QDs in both layers. The preferential nucleation of QDs above underlying QDs will lead to formation of QDs at reduced InAs coverage (depending on the separation of the QD layers, only 70% of the InAs need to be deposited for island formation compared to single layers [74]): if multiple layers

are grown using the same InAs coverage, QDs in the upper layers will be larger than the underlying QDs [73]. Also, the composition of upper layer QDs is influenced by strain-induced intermixing [75, 76], leading to a blueshift of emission from upper layers. This can be compensated by depositing additional InAs [77] and/or reducing the growth temperature [78]. One method to minimize the variation between QDs in vertically-stacked layers is the indium-flush technique [79], where growth is interrupted during the early stages of capping of a QD layer (typically after 2-3 nm of GaAs overgrowth) and the growth temperature is then raised to desorb any remaining surface indium. GaAs capping is then continued for the required spacer layer thickness before the temperature is again lowered for growth of the next QD layer. This produces stacks of truncated QDs of remarkably uniform height, and although it cannot eliminate strain-induced intermixing effects and is limited to the growth of QDs with short wavelength emission, similar QD pairs can be obtained. Electronic coupling between QD pairs can be controlled at the growth stage by altering the separation between them [80, 81, 82] and also by applying an electric field across the QD pair, for example by placing them in a Schottky diode structure [83, 84, 85].

4.3 Spin generation and detection

Introduction of spin polarized carrier populations into semiconductors can be achieved by either electrical or optical injection. Considerable research interest has focused on electrical injection from magnetic contacts [86] or through layers of magnetic semiconductor [87, 88] and the degree of electrical spin injection and spin lifetimes can be evaluated using light-emitting diode (spin-LED) structures, where the polarization of the emitted light is used to determine the spin polarization of electrons and holes recombining in the active region. Several groups have demonstrated QD-based spin-LEDs and have used them to determine the overall spin relaxation time in the device structures, taking into account spin relaxation during carrier transport through the device and capture then recombination in the QDs [89, 90, 91, 92]. However for fast initialization, manipulation and readout of spins for QIP applications, optical methods are preferred. The selection rules for optical transitions near the Γ point in GaAs, arising due to conservation of angular momentum, are illustrated in figure 4.4. The conduction band and valence band states have an angular momentum projection $J_z = \pm 1/2$ for the conduction band electrons, $J_z = \pm 3/2$ for the heavy holes and $J_z = \pm 1/2$ for the light holes. In bulk GaAs at the Γ point, heavy and light holes are degenerate, but because of the difference in transition probabilities from heavy hole or light hole states (a ratio of 3:1 respectively), a maximum of 50% net spin polarization can be generated by excitation with circularly polarized light (figure 4.4(b)). Equivalent selection rules apply to emission (figure 4.4(c)). However, if strain or quantum confinement is introduced, the degeneracy of the hole states can be lifted (figure 4.4(d)), allowing transitions involving only one hole species to be addressed, so that 100% spin polarization can be achieved. Again, equivalent selec-

tion rules also apply for radiative emission, so the spin polarization of carriers can be determined from the circular polarization of the light emitted. This has implications for optical excitation in QD structures: excitation into the GaAs barrier will provide a maximum of 50% spin polarization but excitation into the WL or QD states may provide significantly more spin polarization.

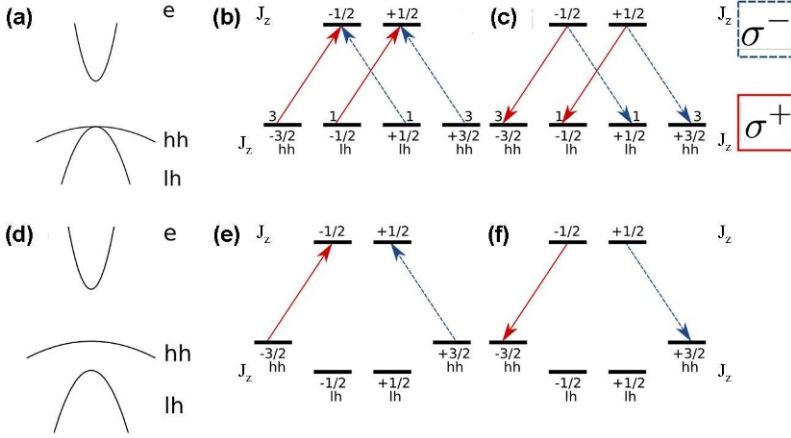


Fig. 4.4 (a) Schematic illustration of bulk III-V band structure, (b-c) selection rules for absorption (b) and emission (c) of circularly polarized light in bulk III-V semiconductor. The numbers by the arrows represent the relative transition strengths. (d) band structure of a confined or strained III-V nanostructure, (e-f) corresponding selection rules for absorption (e) or emission (f) of circularly polarized light in the nanostructure.

The fidelity of these selection rules allows us to measure the spin population in QDs (and other III-V semiconductor-based structures) from the polarization of the emitted light, P_{circ} , defined as:

$$P_{circ} = \frac{I(\sigma^+) - I(\sigma^-)}{I(\sigma^+) + I(\sigma^-)} \quad (4.3)$$

where $I(\sigma^\pm)$ is the intensity of σ^+ or σ^- polarized light, as appropriate. This will depend on the extent of any mixing of the heavy and light hole states. Experimental studies and $\mathbf{k} \cdot \mathbf{p}$ modeling of hole states in InAs/GaAs QDs suggest that they have mainly heavy hole character [93, 94, 95, 96, 97], although other reports have suggested some mixing of hole states due to in-plane anisotropy or strain effects [98, 99, 100, 101, 102, 103]. It is also dependent on the direction of emission and is most reliable in the growth direction [96]. Alternatively, measurements of the spin polarization can be made by the related phenomena of Kerr [104, 105] or Faraday rotation [106, 107, 108, 109]. For these methods, the rotation in polarization of a reflected (Kerr rotation) or transmitted (Faraday rotation) laser beam gives a measure of the magnetization (and hence spin orientation) in a sample, and has been refined for measurements of single QDs [104, 109]. Loss of spin information is

typically described using the characteristic decay times T_1 and T_2 [110]. The T_1 time is the longitudinal spin relaxation time, which relates to the time required for the spin ensemble to reach thermal equilibrium via spin-flip events. If we consider a QD sample excited with circularly polarized light in the growth direction and with a magnetic field also applied in the growth direction (the Faraday geometry), T_1 could be measured considering spin relaxation from the upper to lower Zeeman-split levels. T_2 is known as the transverse spin relaxation time or the spin decoherence time, and gives a measure of the time for coherent precession of the spin before some scattering event changes its phase. Considering the QD sample, if we now apply the field perpendicular to the growth direction (the Voigt geometry) we could make measurements to determine T_2 . For an ensemble of spins, each with potentially different precession and decoherence times, an overall spin dephasing time T_2^* is defined. T_1 and T_2 are related such that $T_2 \leq 2T_1$. The majority of measurements quoting the spin lifetime are of the spin relaxation time T_1 , but for QIP applications we are also concerned with possible loss of coherence of a qubit, so T_2 is relevant. For bulk or quantum well structures, the dominant spin relaxation mechanisms arise due to motion of carriers through the crystal. The D'yakonov-Perel mechanism occurs due to a lack of inversion symmetry in III-V semiconductors which leads to a spin-splitting of the electron states. This spin splitting behaves as a $\mathbf{k}$ -dependent effective magnetic field around which the electron spin may precess. The other dominant spin relaxation mechanism is the Elliott-Yafet mechanism, which describes changes in momentum due to scattering, for example by phonons or impurities. The spin relaxation times for holes in bulk III-V semiconductors are extremely rapid due to mixing of states, and are typically on the order of ps [111]. However, T_2^* times on the order of 100 ps and up to 100 ns for optimized conditions have been measured for electrons in bulk GaAs [112]. For localized carriers in QDs the spin loss mechanisms that are dominant in bulk will be suppressed and even larger spin lifetimes may be obtained, as described in the next section.

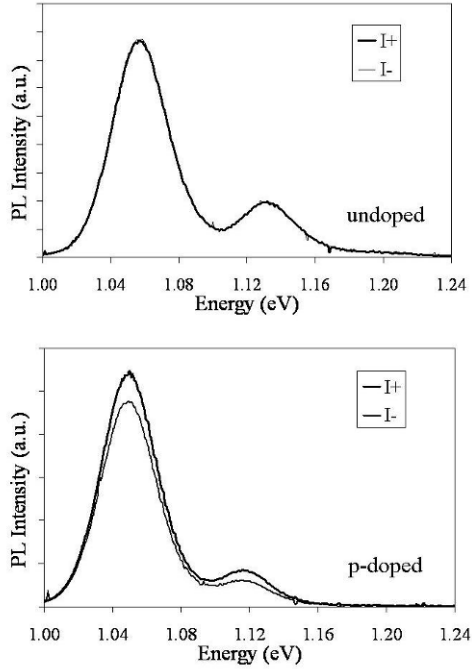
4.4 Spin relaxation and dephasing mechanisms in QDs

Suppression of the D'yakonov-Perel and Elliott-Yafet spin relaxation mechanisms by the localization of carriers in QDs should lead to an extension in spin lifetime. Studies have shown an exciton spin lifetime ranging from a few hundred ps up to around 1 ns [91, 113, 114, 115]. However spin loss mechanisms do remain, and can be enhanced in QDs. In this section we concentrate on the two dominant mechanisms: the electron-hole exchange interaction and the hyperfine interaction. Electron and hole spins can couple by the exchange interaction [116, 117], which occurs in bulk III-V semiconductors but is strongly enhanced by confinement [118] and by asymmetry of the confinement potential [119]. The strong confinement of QDs and elongation of QDs in the $[\bar{1}10]$ direction that reduces the symmetry of the QD from C_{4v} to C_{2v} particularly enhances the asymmetric exchange interaction and this mixes the two bright exciton states $|+1\rangle$, $|-1\rangle$, which give circularly polarized emission,

into states $\frac{1}{\sqrt{2}}(|+1\rangle \pm |-1\rangle)$, which are split by the exchange energy δ_{FS} and are linearly polarized parallel and perpendicular to (110) [117, 120]. An applied magnetic field can be used to distinguish these states: a magnetic field in the Faraday geometry unmixes the spin states and introduces a Zeeman splitting, restoring the circularly polarized $|+1\rangle$ and $|-1\rangle$ states, energetically separated by $g_X \mu_B B$, where g_X is the exciton g-factor, μ_B is the Bohr magneton and B is the magnitude of the applied field [117, 121]. A magnetic field in the Voigt geometry mixes the bright and dark ($|+2\rangle$, $|-2\rangle$) excitons, leading to the observation of a quadruplet rather than a doublet [103, 117, 122]. Paillard *et al.* [121] considered the polarization dependence of time-resolved PL obtained from InAs/GaAs QDs following resonant excitation. For zero applied magnetic field, the linear polarization of the mixed states is long-lived but once a 2.5 T magnetic field is applied in the growth direction to unmix the states, long-lived circular polarization of the PL is observed. This indicated that the exchange interaction results in long-lived mixed states but short-lived pure spin-up or spin-down states. Subsequent studies have confirmed this by demonstrating quantum beats between the mixed states [123, 124, 125, 126] and a correlation has been observed between the magnitude of the fine structure splitting, δ_{FS} , and the exciton spin relaxation rate in single InAs/GaAs QDs [127]. The exchange interaction can be counteracted by doping the QDs. For the X^+ trion state consisting of one electron and two holes, the coupling terms in the Hamiltonian introduced by the exchange interaction between the electron and the spin-up hole are cancelled out by the interaction between the electron and the spin-down hole, so that when exciton recombination occurs it will involve a pure spin state $|+1\rangle$, $|-1\rangle$ resulting in circularly polarized emission (an equivalent argument applies to the X^- trion). P -doping is also expected to enhance the circular polarization of the QD emission, especially for non-resonant excitation into the GaAs matrix, due to faster carrier capture and relaxation times [128, 129, 130, 131], a decrease in the luminescence lifetime of the exciton in p -doped QDs [132, 133, 134], as well as an increased spin lifetime [92, 129, 135].

The extension of the exciton spin lifetime in p -doped QDs can be sufficient for it to exceed the luminescence lifetime, so that circular polarization of emission can be observed even in cw PL spectroscopy, as shown in figure 4.5. Following σ^+ polarized emission into the GaAs barrier, PL spectra analyzed for σ^+ or σ^- polarized light is obtained from both an undoped InAs/GaAs QD ensemble and from a similar ensemble modulation doped at a level of 10 acceptors per QD. For the undoped sample there is no discernable difference between the PL spectra analyzed for σ^+ or σ^- light, but in the p -doped sample there is a clear enhancement of the σ^+ polarized emission, corresponding to a circular polarization $P_{circ} = +5\%$ at the ground state emission peak and $P_{circ} = +15\%$ at the first excited state emission peak. Time-resolved PL measurements obtained from the doped sample show a spin lifetime of 3.6 ns, compared to a luminescence lifetime of 700 ps. We attribute the increased P_{circ} observed from the excited state emission to Pauli spin blocking [136]. For n -doped QDs, a surprising negative circular polarization (NCP) of emission is observed following non-resonant excitation [104, 137, 138, 139, 140, 141, 142, 143]. Polarization-dependent time-resolved PL measurements show a positive circular po-

Fig. 4.5 10 K cw polarization-resolved PL spectra obtained from an undoped QD ensemble and a p -doped QD ensemble with a doping level of 10 acceptors per QD, following excitation into the GaAs barrier with σ^+ circularly polarized light. Emission is resolved for σ^+ and σ^- polarized light, denoted by I+ and I- respectively. Clear circular polarization of emission is observed from the p -doped sample.



larization of emission at short timescales (<20 ps) which then undergoes a rapid reversal, with an increase in NCP over a timescale of 1 ns, after which it decays [137]. This was explained by Cortez *et al.* [137] using the following model. We consider a doping level corresponding to one resident electron per QD and initially the resident spin population is unpolarized. Assuming for simplicity that excitation into the WL with σ^+ polarized light photogenerates only spin-down electrons and spin-up holes and that the electron spin lifetime is sufficiently long to be preserved on capture into the QD while hole spin is randomized, capture of an electron-hole pair by a QD with a single resident electron will result in one of the four equally-likely permutations of carrier occupation illustrated in figure 4.6.

For the examples of figure 4.6(a) and (b), the electron spins are co-polarized and Pauli blocking prevents relaxation to the ground state, whereas for the cases (c) and (d) the electrons have opposite spin so can relax to the ground state to form a spin singlet. States (a), (c) and (d) are bright and can undergo radiative recombination. For (c) and (d), recombination will give σ^+ and σ^- light respectively and will leave a spin-up (c) or spin-down (d) electron. Recombination in case (a) will give σ^+ light and the remaining spin-down electron will rapidly relax to the ground state. However state (b) is dark: it cannot relax radiatively due to conservation of angular momentum. There are two processes by which a Pauli-blocked electron in the excited state can relax to the ground state: a spin flip-flop process where an electron and hole undergo a mutual spin flip and the unblocked electron relaxes to

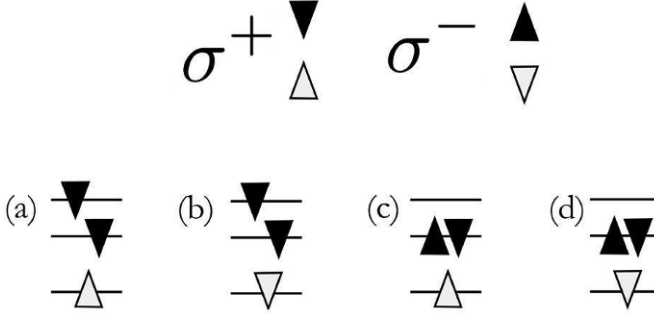
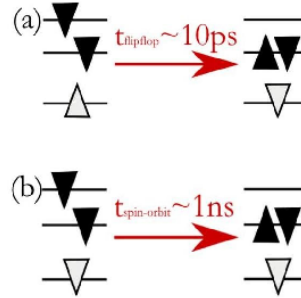


Fig. 4.6 Top: excitation into the WL generates specific excitons depending on the circular polarization of the light, with σ^+ light generating a spin-down electron and spin-up hole and σ^- light generating a spin-up electron and spin-down hole. Equivalent selection rules apply for light emission. Bottom: the four possible trions resulting from capture of a σ^+ photogenerated electron-hole pair (with spin-down electron) by a QD containing a resident electron.

the ground state, and a spin-orbit process where the spin of the electron in the excited state flips due to spin-orbit coupling. These processes are illustrated in figure 4.7, and relaxation of the excited trion state will occur on very different characteristic timescales depending on its configuration. For a bright state (figure 4.7(a)), the spin flip-flop process is extremely efficient, occurring on a timescale, $t_{flipflop}$, of around 10 ps. For the dark state, the asymmetric exchange interaction is much smaller and so the flip-flop mechanism is suppressed, so the dark exciton will relax via the spin-orbit interaction, occurring over a much longer timescale, $t_{spin-orbit} \sim 1$ ns.

Fig. 4.7 (a) relaxation of the bright excited trion by an efficient electron-hole flip-flop process with a characteristic time of 10 ps, (b) relaxation of the dark excited trion by the spin-orbit interaction with a characteristic time of 1 ns.



We now consider the time evolution of the circular polarization of the PL emission. The four possible exciton combinations shown in figure 4.6 are shown at three representative times: $t < t_{flipflop}$ (figure 4.8), $t \sim t_{flipflop}$ (figure 4.9) and $t \sim t_{spin-orbit}$ (figure 4.10). At very short times after excitation ($t < t_{flipflop}$), the recombination possibilities for the four exciton combinations are as described above, and this leads to a positive net circular polarization (+33%), with the remaining electrons predominantly spin-down. For $t \sim t_{flipflop}$, the behavior of states (b), (c)

and (d) remain the same but state (a) can relax by the flip-flop process. In this case the mutual spin flip leaves a bright exciton which recombines giving σ^- light and a spin-down electron remaining. Now the overall net circular polarization will be negative (-33%). Once we reach times $t \sim t_{spin-orbit}$, relaxation of the dark exciton (b) can occur by the spin-orbit interaction and the resulting singlet state can recombine to give σ^- light, leaving a spin-down resident electron, so the overall net circular polarization will now be -50% and the remaining electrons will be majority spin-down.

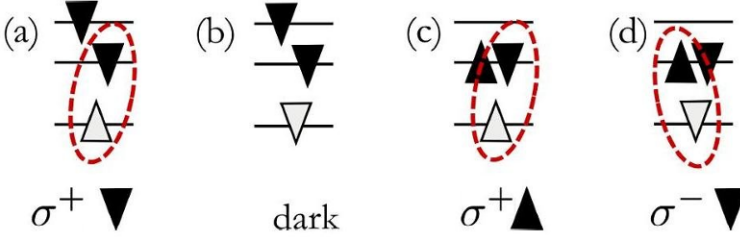


Fig. 4.8 The four possible trion states at time $t < t_{flipflop}$, with allowed transitions for radiative recombination highlighted by dashed ovals and the circular polarization of the emitted light and spin-polarization of the remaining electron shown below. Net P_{circ} of the emission is $(2 - 1)/(2 + 1) = 33\%$ and the remaining electrons are majority polarized spin-down.

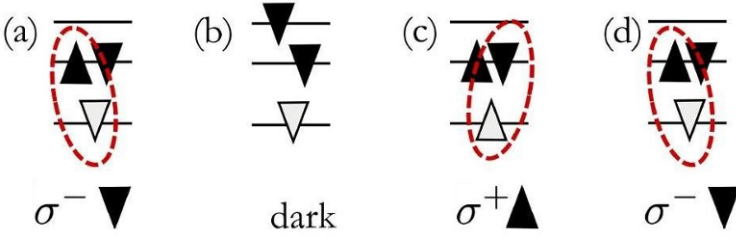


Fig. 4.9 The four possible trion states at time $t \sim t_{flipflop}$. The bright excited trion (a) can relax via the electron-hole flip-flop process. Net P_{circ} is $(1 - 2)/(2 + 1) = -33\%$ and the remaining electrons are majority polarized spin-down.

The model qualitatively reproduces the dynamics of the circular polarization and suggests that circular polarized light can write a spin to the resident electron in a QD. Pump-probe experiments using a circularly-polarized pump and linearly-polarized probe pulses have been used to determine a spin lifetime of the resident electron in n -doped QDs of 15 ns [137]. NCP appears to be a general feature of n -doped QDs [139] but disappears at high n -doping [135]. However the extension of the spin lifetime of single resident carriers in a QD to far beyond the radiative lifetime of excitons in a QD allows for the implementation of spin memory using single carri-

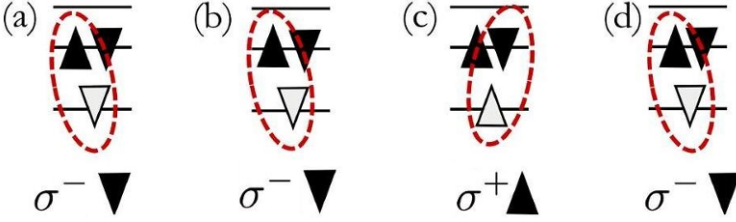


Fig. 4.10 The four possible trion states at time $t \sim t_{\text{spin-orbit}}$. The dark excited trion (b) can relax via the spin-orbit interaction. Net P_{circ} is $(1 - 3)/(3 + 1) = -50\%$ and the remaining electrons are majority polarized spin-down.

ers that can be optically initialized and read out. For single carriers in a QD at low temperatures, the dominant remaining spin relaxation mechanism is the hyperfine interaction with surrounding nuclear spins. All nuclear species in a III-V semiconductor will have a non-zero nuclear spin, and an electron confined in a QD will be coupled to 10^4 - 10^6 nuclear spins. Following Coish and Baugh [144], the Hamiltonian for an ensemble of nuclear spins can be written as follows:

$$H = H_Z + H_{hf} + H_{orb} + H_{dd} + H_q \quad (4.4)$$

where H_Z is the Zeeman energy in an applied magnetic field, H_{hf} is the hyperfine term comprised of two parts $H_{hf} = H_c + H_a$, where H_c is the contact hyperfine interaction and H_a is the anisotropic hyperfine interaction, H_{orb} is a coupling term between the nuclear spin and electron orbital angular momentum, H_{dd} is the dipole-dipole interaction between nuclear spins and H_q is the quadrupolar interaction between nuclear spins and an electric field gradient. When considering the coherence of an electron spin in a QD, the contact hyperfine term is the most important. Over short timescales compared to the nuclear spin precession time ($\ll 1\mu\text{s}$), we can consider the nuclear spins to be frozen and their random alignment will lead to an effective magnetic field, known as the Overhauser field, which is generally on the order of a few mT and about which the electron spin can precess [145]. This randomly aligned Overhauser field will lead to a rapid decay in polarization of an electron spin initially polarized in the growth direction, to $1/3$ of its initial value over a ns timescale, but this decay can be suppressed by a moderate external magnetic field (>100 mT) [146]. Under high magnetic fields (>4 T), exceptionally long spin relaxation times (T_1) of around 1 ms have been reported [147, 148]. Decoherence of the electron spin (T_2) occurs as a result of non-uniformity of the hyperfine interaction due to the spatial variation of the electron wavefunction in the QD [149] and can occur on a ns timescale. To extend the coherence time of the electron, alignment of the nuclear spins along the direction of polarization of the electron spin is required. The hyperfine interaction itself provides one method for achieving this: by optically pumping the QD with circularly polarized light to write a polarization to the resident electron spin, this spin polarization can be transferred via the hyperfine interaction to the nuclear spin ensemble, a process known as dynamical nuclear polarization

(DNP) [150]. This offers us the opportunity to establish highly localized magnetic fields in particular QDs, on the order of a few tesla [151], which can then be used for manipulation of electron spins. The nuclear ensemble can also be used in this way as a long-lived spin memory [152], since the dephasing time of the nuclear spin ensemble will be on the order of μs , limited by dipole-dipole interactions between nuclear spins. The nuclear spin dephasing time can be further extended by removing the resident electron after optical pumping, for example by an applied bias for a QD in a Schottky diode structure [153], and controllable switching of the nuclear spin polarization by DNP has been demonstrated [154]. However in order to significantly extend the coherence time of the electron spin, almost complete polarization of the nuclear spin ensemble is required [155], and for In(Ga)As/GaAs QDs, the maximum nuclear spin polarization currently achieved via DNP is only around 50% [156, 157], possibly limited by a reduction in the efficiency of DNP due to electron Zeeman splitting arising from the Overhauser field or an applied magnetic field or by diffusion of nuclear spins [158, 159]. Fortunately, an increase in the electron spin coherence time can also be achieved by narrowing the distribution of nuclear spins using a variety of strategies [160, 161, 162, 163, 164, 165, 166], and in some reports this has extended the electron T_2 time towards 1 μs , comparable with the nuclear spin precession time. Alternatively we can use hole spins rather than electron spins as the basis of a qubit. Unlike in bulk, where mixing of hole states results in rapid spin relaxation, holes confined in QDs are expected to maintain their spin for a time that is comparable with spin relaxation times of electrons. Initialization of single hole spins using circularly polarized light has been demonstrated [167] and similar T_1 times for holes and electrons (up to 270 μs) have been measured in InGaAs QDs with an applied magnetic field of 1.5 T [97], with multiple phonon processes and spin-orbit interaction expected to be the dominant spin relaxation mechanisms [168]. However because of the p-orbital nature of valence band states, the hole wavefunction is zero at the location of nuclei and so the contact hyperfine term is suppressed. The hyperfine interaction is then determined by the anisotropic hyperfine term, H_a , and the coupling of orbital angular momentum to the nuclear spin, H_{orb} [169]. This results in the hyperfine interaction between the hole spin and nuclei being an order of magnitude less than for electron spins [170] and coherence times in excess of those achievable with electron spins are a possibility. This has been demonstrated in a recent experiment [171] that probes the time-averaged coherence time (T_2^*) of a single hole spin in an InGaAs QD, indicating $T_2^* > 100$ ns, with a 40% likelihood that T_2 exceeds 1 μs (the uncertainty due to the resolution of the spectroscopy).

4.5 Outlook

Recent work has demonstrated the viability of electron or hole spins localized in QDs as a basis for constructing qubits for QIP, with the coherence times of these spins on the order of μs . Using optical pulses, ultrafast initialization of the spin

states at ps timescales has been reported [103, 172] and now the focus of research is moving to the manipulation of the spins and formulation of quantum operations. By resonant excitation of particular states, Rabi oscillations demonstrating the controlled rotation of spin has been observed and this can be made conditional on the number or spin state of carriers in a single QD [173] or coupling in a QD molecule [174]. Manipulation of spins in this way can be carried out using ultrafast optical pulses on the order of ps [175, 176, 177], so protocols for operation of robust quantum gates can be designed, with the possibility of carrying out a sufficient number of operations on the qubit before coherence is lost for implementation of practical QIP schemes. Much of this work is still in its infancy and many significant technological challenges remain, for example increasing the number of interacting qubits, but the progress made so far, in little over a decade since the first proposal for quantum computation using quantum spins [178], is highly encouraging.

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Chapter 5

Optically induced spin rotations in quantum dots

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Abstract The spin of an electron trapped in a quantum dot is currently of interest both because it constitutes a prototype quantum mechanical system in a controllable solid state environment and due to its relevance in quantum information processing. For such applications, a high level control of the spin is necessary, and various techniques from time dependent magnetic and electric fields to lasers are used. Here we develop the basic ideas involving the quantum dot electron spin for quantum information applications and review optical methods of its control. Particular emphasis is given on the use of hyperbolic secant optical pulses. Relevant experimental results are also briefly discussed.

5.1 Introduction

Spins in quantum dots (QDs) are attractive candidates for qubits [2] in quantum information processing. Semiconductor materials are typically used in information technologies, and spin coherence times can be very long in semiconductor QDs [4, 11] compared to the times in higher dimensional structures. Most importantly they are long compared to the typical time for quantum control. In the last decade there has been much research on growing QDs, on understanding their physics, and on designing and implementing their quantum coherent control. Aside from use as qubits, they are also excellent photon emitters due to their large dipole moments as compared to atoms. Of course the large dipoles also mean faster optical decay rates, an issue that must be addressed in optical control. Their differences from atomic

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systems stem from the underlying solid state material, their size and the resulting tunability. The interplay of the solid state nature of these structures and the atomic-like character of their sharp levels makes this field an interesting arena in which to study quantum mechanical effects.

Among the qubit requirements for quantum computation (the so-called ‘Di Vincenzo criteria’ [5]) is the ability to implement arbitrary unitary single-qubit operations. An arbitrary unitary transformation of a spin (or any two-level system) can be viewed as a rotation of the spin (or pseudospin). So, the requirement of unitary single spin control is equivalent to the requirement of arbitrary spin rotations. In practice this means that given a two-level system and its stationary Hamiltonian H_o , we need to find the fields which will couple to the system through a time-dependent Hamiltonian $H_c(t)$ (the subscript c is for ‘control’) such that the evolution operator U , which is a solution of $HU = i\dot{U}$, is the target rotation and $H = H_o + H_c$. Note that the design must be for U itself and not for the final state, which is generally unknown, so that designing an arbitrary U is more general than finding a way to transfer population between two quantum states. To clarify this point, consider a rotation $R_x(\pi)$ ¹ acting on the spin ‘down’ eigenstate, $|\bar{z}\rangle$. The final state is the ‘up’ eigenstate. This is an example of population transfer (in the sense that initial and final populations reside in energy eigenstates), which could be achieved by an infinite number of rotation operators $R_{n_\perp}(\pi)$, where $n_\perp$ is any axis in the xy plane. However, these rotations are not equivalent. If the initial state was instead a superposition of the two eigenstates, for example $|z\rangle + i|\bar{z}\rangle$, then $R_x(\pi)$ would take it to $|z\rangle - i|\bar{z}\rangle$. All the other operations $R_{n'_\perp}(\pi)$, where $n'_\perp \neq x$ change the state in different ways. One can see from this that testing the effect of an operator on initial states pointing along orthogonal directions on the Bloch sphere reveals information about these operations that a mere application on one state would not give. This is basically the idea of quantum process tomography [6, 7].

The QDs we will focus on here are called ‘self-assembled’ quantum dots and are, as the name implies, spontaneously formed islands of one semiconductor material on top of another during growth by Molecular Beam Epitaxy. Typically such QDs are InAs (or InGaAs) on a GaAs substrate and overgrown by GaAs. These QDs tend to grow in pyramid shape due to strain, though techniques exist to truncate their tops and make their properties more homogeneous. Their sizes are in the few nanometer scale (height 2-5 nm and base 10-20 nm). Another semiconductor structure, which is also called a QD, is an electrostatically defined region on a quantum well, where electrons are trapped by gates. They have also been called ‘transport quantum dots’. These QDs are about an order of magnitude larger than the self-assembled ones, and thus their energy levels are much closer to one another. These are controlled by rf magnetic and electric fields. We briefly comment on some main ideas and experiments on their manipulation below.

This Chapter is organized as follows: In Section 5.2 we will review some basic concepts, namely the spin vector and composite rotations. Section 5.3 contains a brief overview of some important developments in the rf control of the spin in QDs. The

¹ The notation we use for a rotation operator is $R_n(\phi)$, where n and ϕ denote the axis and angle of rotation respectively.

main Section in this Chapter is 5.4, which deals with optically induced spin rotations. The selection of the presented work is by no means comprehensive. It has been chosen to highlight (a) the various physical phenomena that are used in optical control, such as, e.g., Optical Stark effect and Coherent Population Trapping, and (b) some recent prominent experiments demonstrating spin rotations.

5.2 Useful concepts

It is useful to introduce some general concepts and ideas first. Here we describe the idea of the spin vector and the concept of composite spin rotations. We also explain the phenomenon of coherent population trapping as well as the concept of fidelity in Appendix 5.6.1.

5.2.1 Spin state as vector on Bloch sphere

A spin state, or any pure state of a two-level quantum system $\{|0\rangle, |1\rangle\}$ (a qubit), can be written as

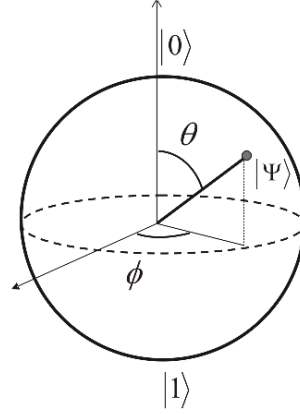
$$|\psi\rangle = \cos \frac{\theta}{2} |0\rangle + e^{i\phi} \sin \frac{\theta}{2} |1\rangle, \quad (5.1)$$

so that it can be parameterized uniquely by two angles, a polar ($0 \leq \theta \leq \pi$) and an azimuthal ($0 \leq \phi < 2\pi$), as shown in Fig. 5.1. This means that it can be represented as a vector on a sphere with radius unity. The components are given by $s_j = \langle \psi | \sigma_j | \psi \rangle$, where the σ_j ($j = x, y, z$) operators are the Pauli spin matrices. The sphere is the well-known Bloch sphere, and the spin is called Bloch or spin vector. The beauty of this representation is that it allows the classical view of a vector rotating in three-dimensional space. Moreover, it can be easily shown that (i) any mixed state ρ can also be thought of as a vector with components $s_j = \text{Tr}(\rho \sigma_j)$, of length less than unity, which is therefore inside the sphere and (ii) any unitary operation can be expressed as a rotation of the spin vector.

5.2.2 Composite rotations

It is obvious that the application of two (or more) rotations is itself a rotation, since the product of two unitaries is also a unitary and any unitary operation of a qubit can be represented as a rotation. A more general way to view this is that since rotations form a group, a product of any two elements of the group is itself a group element. Composite rotations are of interest both because in some cases rotations about only a certain set of axes are experimentally achievable, and also because composite ro-

Fig. 5.1 Bloch sphere representation of the spin. The qubit state is parameterized by an azimuthal and a polar angle. Any unitary operation on the qubit can be represented as a rotation of the spin vector.



tations can reduce systematic errors when compared to a ‘single-shot’ rotation [1]. The topic of pulse sequence design for implementing composite rotations has been extensively studied in NMR, where most of the work was in the context of population transfer (or ‘magnetization inversion’); for a review see [8]. These ideas have been borrowed and expanded for quantum operations in the context of quantum information processing [9].

5.3 rf control of spin in quantum dots

For an electron spin trapped in a quantum dot in a magnetic field $\mathbf{B} = B\mathbf{z}$, the most obvious way to implement spin rotations is by a time-dependent magnetic field in the xy plane, which oscillates near or at the resonant frequency, i.e., the energy separation of the two states. This is electron spin resonance (ESR). For a square pulse, the well-known analytic solution is due to Rabi [29]. The oscillatory magnetic field, which is on for time τ , is applied to the spin. The evolution operator in the interaction picture (laser frame) then has the form:

$$U = \begin{pmatrix} \cos\left(\frac{\tau\Omega_0}{2}\right) + i\frac{\Delta}{\Omega_0}\sin\left(\frac{\tau\Omega_0}{2}\right) & -2i\frac{\Omega}{\Omega_0}\sin\left(\frac{\tau\Omega_0}{2}\right) \\ -2i\frac{\Omega}{\Omega_0}\sin\left(\frac{\tau\Omega_0}{2}\right) & \cos\left(\frac{\tau\Omega_0}{2}\right) - i\frac{\Delta}{\Omega_0}\sin\left(\frac{\tau\Omega_0}{2}\right) \end{pmatrix}, \quad (5.2)$$

where Ω and Δ are the Rabi frequency and detuning respectively, and $\Omega_0 = \sqrt{\Delta^2 + 4\Omega^2}$ is the so-called grand or effective Rabi frequency. Note that when $\Delta = 0$ the rotations are about an axis in the xy plane², while for $\Delta \neq 0$ the axis of rotation has a z component. Composite rotations can give a net rotation about the quantiza-

² Actually the evolution operator as written in Eq. (5.2) expresses rotations about the x axis, which is arbitrarily chosen. Rotations about other axes in the xy plane can be obtained by adjusting the phase between subsequent pulses.

tion axis or other axes. This kind of rotation has been demonstrated experimentally in ‘transport’ quantum dots [10].

Although conceptually straightforward, this kind of control has some inherent shortcomings that optical control can in principle circumvent. Typical Zeeman splittings are on the order of μeV , which means that RF magnetic fields are used for control. Such fields have envelopes that are on the order of nanoseconds, setting an upper limit in the speed of the control. This value is to be compared to the spin coherence time, which has been measured [4] to be on the order of μsec . In addition, RF magnetic fields are not spatially focused³, so that when scaling up it will be extremely challenging to focus down to a single spin and not affect neighboring qubits. A time varying magnetic field is accompanied by electric fields, which couple to the orbital states of the QD. In [10] additional fields were used to compensate for such the unwanted electric fields. However, in a multi-qubit setting this would require a large increase in overhead.

Another approach to spin rotation that has been proposed theoretically [24, 25] and recently demonstrated experimentally [39] is to use time-dependent electric fields, which can be spatially more focused as they are generated on chip. Since the electric field does not couple directly to the electron spin, the interaction is mediated by spin-orbit coupling. Such electrical control, however, is still rather slow, on the order of tens of nsec.

5.4 Optical control of spin in quantum dots

The challenges mentioned above can be overcome by use of optical control. The electric field of a laser does not couple directly to the spin, but instead couples to an interband transition of the quantum dot, so this is also a spin-orbit mediated coupling. The laser promotes an electron from the valence to the conduction band, leaving behind a hole. The resulting electron-hole pair is known as an exciton and along with the resident extra electron in the QD forms a three-particle state called a trion. The goal in optical approaches is to use this auxiliary optically excited state to implement a rotation of the spin. The advantages of optical control are that laser pulses are fast, on the scale of a few picoseconds, and they can be spectrally and spatially focused. The spectral selectivity is important: the various QDs in the sample have different band gap energies determined by their size, composition, etc. This gives us a handle on addressing a QD selectively without affecting other QDs since their transition energies can be determined via spectroscopy. An additional advantage to using optical transitions is that they provide a natural way of coupling the spin qubits to photons, which are flying qubits and can transmit information between spins in spatially separated QDs.

³ The QDs have the same or very similar Zeeman splittings, so spectral selectivity cannot be used as a tool either.

5.4.1 Energy levels and selection rules

Let us first look at the energy levels in the quantum dot. Due to quantum confinement, the single-particle levels become sharp. The QD is small by macroscopic standards, on the order of nm, but it is large compared to typical interatomic distance in the solid, which are in the Å range. This means that a large number of atoms are included in the QD, so that some features of the solid are still present. The state of the electron can then be expressed as a Bloch function multiplied by an envelope function which has a typical length scale the size of the QD, while the variation of the Bloch functions is on the atomic scale. This theoretical treatment is similar to that of impurities in semiconductors [30]. The III-V dots in which we are interested have a direct band gap, with the top of the valence band originating from atomic p orbitals and the bottom of the conduction band having s character. Due to spin-orbit interaction the states in the valence band are split according to the value of total angular momentum J in the bulk semiconductor, the top of the valence band having $J = 3/2$. The $J = 1/2$ doublet is on the order of 0.1–0.5 eV lower in the semiconductors of interest here and is called ‘split-off’ hole band. In quantum wells and quantum dots, where the symmetry of the system has been broken, there is additional splitting—this time within the $J = 3/2$ states. The $J = 3/2$ states with projection $M_J = \pm 1/2$ are deeper in energy by about 20–30 meV compared to those with $M_J = \pm 3/2$. The former are called light hole states and the latter heavy hole states. In QDs light hole states have not been seen in spectroscopy experiments, but their effect can be taken into account perturbatively. The excitation used for optical control is the lowest energy one, a transition which involves the heavy hole. The absence of the light holes from that region of the spectrum is advantageous because it simplifies the number of transitions and their selection rules.

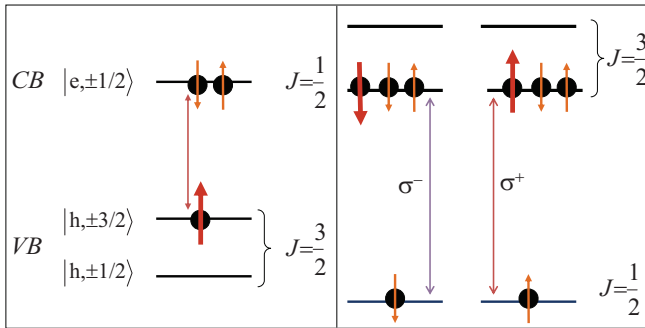


Fig. 5.2 Single particle levels in a QD are shown in the left panel. The bands of the bulk semiconductor have split into sharp, atomic-like states. A single resident electron in the conduction band is depicted, as well as an optically generated exciton. The thin (thick) line represents the electron (hole) spin. In the right panel the total energy states (i.e., Coulomb interactions taken into account) along with the selection rules can be seen. The ground states are electron spin states polarized along the growth axis (z) and are labeled accordingly $|\uparrow_z\rangle, |\downarrow_z\rangle$, and the excited states are the trions pointing up also along the z axis. We will focus on the heavy hole trions and label them $|T\uparrow\rangle, |T\downarrow\rangle$.

The single-particle QD levels are pictured in the left panel of Fig. 5.2. This depiction, though familiar to solid state physicists, is inaccurate for the description of the bound states such as excitons and trions. When taking into account the Coulomb interactions between the particles, the total energy level structure is the proper picture to use, as shown at the right panel of Fig. 5.2, where the selection rules for circularly polarized light propagating along the growth axis are also shown.

5.4.2 Optical spin rotations

In the geometry of Fig. 5.2 the only rotations one can obtain are about the z axis. This is because rotations about axes other than z will necessarily transfer population between the $|z\rangle$ and $|\bar{z}\rangle$ states. But in Fig. 5.2 there are no optical transitions coupling those two states. Since we need rotations about at least one more axis, we need to enable those (indirect) transitions. This can be done by switching on a magnetic field in the plane of the QD (perpendicular to the optical axis—a parallel field would not change the symmetries in Fig. 5.2). Then the B field defines the x axis and mixes the $|\pm z\rangle$ states into the new energy eigenstates. The effect of the magnetic field along the x axis on the trion states is more subtle. Since the heavy hole states are not directly coupled to each other with a B field but through the light hole states, which are energetically separated by Δ_{hl} , the Zeeman interaction will be rescaled by $(g_h\mu_B B/\Delta_{hl})^2$, where g_h, μ_B are the g factor for the hole and the Bohr magneton respectively. In InAs self assembled QDs of interest here a splitting between the two heavy-hole trions has been observed in spectroscopy [40]. We will treat the two heavy-hole trions as a pseudospin represented by an effective heavy-hole g factor and take its value from experiment.

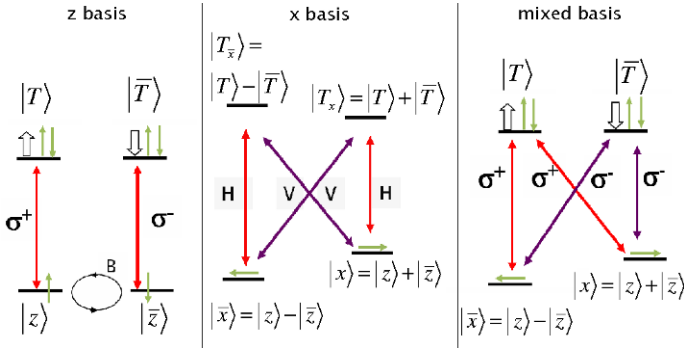


Fig. 5.3 Optical selection rules in three different bases: the z basis, which is the optical axis, where the polarization selection rules are the simplest for circularly polarized light, the x basis, which is the energy eigenbasis, and the ‘mixed’ basis, where the trion is in the z basis, but the electron in the x basis.

It is useful when thinking about control of the spin to have the optical selection rules in mind in different bases. The reason is that due to the geometry, and depending on the optical polarization used, energy and polarization selection rules are simpler in different bases. In the following we will use the energy or x basis, the optical/growth or z basis, and the mixed basis, all shown in Fig. 5.3.

With this energy structure and optical selection rules, the question of spin rotation becomes a mathematical one, and can be posed as the following inverse problem: Find the laser field(s) that will yield an evolution operator that satisfies the Schrödinger equation $HU = i\dot{U}$ with the property $U(t_f) = U_{\text{target}}$, where t_f is the final time⁴. Note that $U(t_f)$ is generally a 4×4 operator. A challenge in optically induced spin control as opposed to ESR is the requirement of returning the entire population to the spin subspace after the action of the pulse, i.e., in the basis $|z\rangle, |\bar{z}\rangle, |T\rangle, |\bar{T}\rangle$ we require:

$$U(t_f) = \begin{pmatrix} U_{2 \times 2} & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}. \quad (5.3)$$

That is, we want to find the fields such that $U_{2 \times 2} = R_n(\phi)$, where n, ϕ parameterize the target rotation.

An additional complication related to the fact that an optically excited auxiliary state is used is that there may be recombination of the optically excited e-h pair accompanied by spontaneous emission of a photon. This is an effect that will lower the fidelity (a measure of how good the operation is, for an exact definition see Appendix 5.6.2) of the rotation and ought to be avoided. In the existing rotation proposals the control is initially designed considering only the unitary dynamics based on the laser fields. In most of the proposals the design is such that spontaneous recombination will not play a large role, for example by using very off resonant pulses and exciting the trion only virtually, or by using ultrafast pulses so that the trion is excited for very short times. In [33, 34] the effect of spontaneous emission on the dynamics was determined by numerically solving the Liouville-von Neumann equations, and the fidelity was calculated.

The time-dependent Schrödinger equation is difficult to solve analytically, even for two-level systems. There are some well-known cases of pulses that can be analytically solved in the case of a two-state system, such as the Rabi problem [29] and the family of the hyperbolic secant pulses [16, 17]. Approximate solutions also exist for adiabatic pulses. In the case of spin rotation where 3 or 4 levels are involved the common approach of all current proposals is to reduce the system to a two-level system by use of appropriate laser fields. The way this is done varies from approach to approach. Below we will review some of the theoretical schemes for optical spin rotation in QDs, with particular emphasis on the one based on the hyperbolic secant pulses [33, 34], which has shown remarkable agreement with recent experiments [22].

⁴ strictly speaking, t_f is infinity but in practice it is the time where the pulse is effectively zero

5.5 Optical spin rotation proposals

5.5.1 Optical Stark effect based rotation

In the optical Stark effect a continuous wave (CW) laser (or square pulse) acts on a transition and causes a shift to the ground state (one of the spin states in our case). To find the value of that shift we transform to the rotating frame of the laser (or, equivalently, the ‘dressed basis’) where the Hamiltonian is time independent. Then, for large detunings (and weak fields), we can use ordinary perturbation theory. To second order, the shift is Ω^2/Δ . When the pulse used is circularly polarized, e.g., σ^+ , only the $|+z\rangle$ spin state has its energy shifted (the ‘ z ’ basis is the most appropriate description for this approach). This is like having an effective magnetic field that splits the two spin states along z , so that it can be used to induce only rotations about the z axis. When the laser is pulsed instead of CW the physics is similar. This method has been used for rotations of spins about the growth axis in quantum wells [27] and more recently in quantum dots [28]. In [28] the combination of spin precession about the external B field and the optical Stark shift was used to induce spin rotations about general axes.

5.5.2 Adiabatic approaches to spin rotation

Another method of spin rotation is based on adiabatic elimination of the trion. When the pulses used are slow enough to satisfy the adiabatic condition the trion state is not excited. Such techniques were first used for population transfer without losses and are known as STIRAP techniques. STIRAP (Stimulated Raman Adiabatic Passage; for a review see [32]) has been adapted to design a spin rotation by several researchers. Here we will look at two proposals, both of which use slow pulses as dictated by an adiabatic condition such that the trion remains unpopulated throughout the operation.

A proposal of Kis and Renzoni [12] uses the original STIRAP for population transfer along with coherent population trapping (CPT—see appendix 5.6.2 for a brief discussion) and laser-induced phases. The idea is to use the original three-level system along with an additional lower state. Then a pair of lasers is used to define the bright/dark (B/D) basis in a CPT scheme and another laser—switched on first—is used to couple the excited state to the auxiliary lower level, $|aux\rangle$. Then traditional STIRAP can be used so that the population from $|B\rangle$ is transferred to $|aux\rangle$. A second application of these pulses, in reversed time order, with lasers that have a phase ϕ relative to the original ones will transfer the population back to $|B\rangle$, thus inducing a phase shift to state $|B\rangle$. This relative phase is a rotation about the axis defined by the B/D basis. Changing the B/D composition and the phase ϕ allows any rotation. This approach has the stringent requirement of an extra lower level, which is not present in a single charged quantum dot. In Ref. [13] it was suggested that a QD molecule

composed of two tunnel-coupled QDs with the population being transferred to the additional QD. This is an extra complication from an experimental point of view, allowing for errors and doubling the number of QDs required.

In another STIRAP-like approach [14] the two transitions are also implicitly assumed to be addressable separately (fast oscillating terms are dropped based on the requirement that the Zeeman frequency is larger than the Rabi frequency), and the two-photon resonance condition is also employed. Now we are using the ‘mixed basis’ from Fig. 5.3. Then, moving to the rotating frame yields a Hamiltonian without oscillating terms in which the whole time dependence is in the envelope of the E -field. The adiabatic approximation then reads

$$|\dot{\xi}| \ll 2\sqrt{\Omega_{\uparrow}^2 + \Omega_{\downarrow}^2 + \left(\frac{\Delta}{2}\right)^2}, \quad (5.4)$$

where $\xi = 1/2 \arctan\left(2\sqrt{\Omega_{\uparrow}^2 + \Omega_{\downarrow}^2}/\Delta\right)$, and $\Omega_{\uparrow(\downarrow)} = \Omega_{\uparrow(\downarrow)}(t)$ is the Rabi frequency acting on the up (down) state. This approximation essentially allows one to treat the Rabi frequency as a constant and the eigenstates of the Hamiltonian are found by solving an eigenvalue equation as if the Hamiltonian were time-independent. The explicit time dependence appears later and is integrated over when finding the induced phases. This is the standard treatment of adiabatic evolution where the system is always in an instantaneous eigenstate of the Hamiltonian. This approach allows all rotations to be generated. Varying the relative intensity and the phases of the two pulses determines the rotation axis, just as in the CPT scheme, while the absolute intensity of the pulse and the detuning determine the rotation angle, which is given by

$$\phi = \int dt \left(\frac{\Delta}{2} - \sqrt{\left(\frac{\Delta}{2}\right)^2 + \Omega_{\uparrow}^2 + \Omega_{\downarrow}^2} \right). \quad (5.5)$$

Because of the requirement in this approach that the Rabi frequency be much less than the precession frequency, for low Zeeman splittings this proposal requires relatively long pulses in order to accumulate large rotation angles [see Eq. (5.5)].

5.5.3 Hyperbolic secant based rotations

A pulse with a temporal hyperbolic secant envelope has some very attractive properties. First, it is one of the few pulse shapes for which the Schrödinger equation is analytically solvable in a two-level system. The solution was first found in the early 1930s by Rosen and Zener [16] in the context of a spin in a time dependent magnetic field. Some decades later it was rediscovered in the context of the phenomenon of self-induced transparency [38], which is the lossless propagation of an optical pulse through an optically dense medium. In the context of optical spin ro-

tations the availability of an analytical solution and some simple properties of the sech pulse are very attractive. First we will briefly review the solution of Rosen and Zener, and then show how the sech pulse can be used for spin control. We will concentrate on the theoretical aspects and include some experimental results. Further data and details on the experimental implementation can be found in [22] and in Chapter 6 of this book.

5.5.3.1 Review of the Rosen-Zener solution

Consider a two-state system with a ground state $|g\rangle$ and an excited state $|e\rangle$, coupled by a time dependent Hamiltonian with a sech envelope and central frequency ω_o . The coupling matrix element in the rotating wave approximation is:

$$\langle g|H(t)|e\rangle = \Omega \operatorname{sech}(\sigma t) e^{i\omega_o t} \equiv f(t) e^{i\omega_o t}, \quad (5.6)$$

where Ω is the Rabi frequency, σ is the bandwidth of the pulse. Then the Schrödinger equation reads

$$H(t) \begin{pmatrix} c_g \\ c_e \end{pmatrix} = i \frac{d}{dt} \begin{pmatrix} c_g \\ c_e \end{pmatrix} \quad (5.7)$$

where

$$H(t) = \begin{pmatrix} 0 & f(t) e^{i\omega_o t} \\ f(t) e^{-i\omega_o t} & \omega \end{pmatrix}. \quad (5.8)$$

Moving to the interaction picture, the problem reduces to solving two coupled first order differential equations or, equivalently, one second order equation of the form:

$$\ddot{c}_e + (i\Delta - \dot{f}/f)\dot{c}_e + f^2 c_e = 0, \quad (5.9)$$

where c_g (c_e) is the amplitude of the ground (excited) state, $\Delta = \omega - \omega_o$ is the detuning, and with the initial condition $c_e(-\infty) = 0$. By a change of variable

$$\zeta = \frac{1}{2} (\tanh(\sigma t) + 1), \quad (5.10)$$

RZ bring the equation into the form of the Hypergeometric equation, where

$$a = \frac{\Omega}{\sigma}, \quad (5.11)$$

$$c = \frac{1}{2} \left(1 + i \frac{\Delta}{\sigma} \right). \quad (5.12)$$

After imposing the initial conditions, the amplitudes of $|g\rangle$ and $|e\rangle$ are, respectively:

$$c_g = F(a, -a, c^*, \zeta) \quad (5.13)$$

$$c_e = -i \frac{a}{c^*} \zeta^{1-c} F(a+1-c, 1-a-c, 2-c, \zeta). \quad (5.14)$$

We see from Eq. (5.14) and by use of the properties of the Hypergeometric function that when

$$\sigma = \Omega \quad (5.15)$$

there is no population transfer to the excited state for $t \rightarrow \infty$, i.e., $c_e(\infty) = 0$. Instead the pseudospin vector undergoes a full cycle from $|g\rangle$ to $|e\rangle$ and back to $|g\rangle$ with the ground state having acquired a phase factor

$$c_g(\infty) = -\frac{\sigma + i\Delta}{\sigma - i\Delta} \equiv e^{-i\phi}, \quad (5.16)$$

$$\tan \phi = \frac{2\sigma\Delta}{\Delta^2 - \sigma^2}. \quad (5.17)$$

For σ fixed, the path will be determined by the detuning. It is remarkable (and in our case useful) that the condition of a full ‘Rabi flop’ is independent of the detuning. This is true for sech pulses even when the pulse envelope is long.

5.5.3.2 Use of RZ pulses for composite spin rotations

Rotations about the z axis

First we consider a small Zeeman splitting in an external magnetic field B along the x (in-plane direction). By ‘small’ we mean as compared to the bandwidth of the optical pulse. In the time domain this is equivalent to a fast pulse compared to the spin precession. We will use σ^+ light (and thus consider the z basis), and review the z rotations as developed in [33]. For an arbitrary *sech* pulse, the evolution operator of the three-level system (the other trion state is decoupled for the polarization used), under the approximation of slow precession, $\omega_B \ll \sigma$, is given by

$$U \simeq \begin{pmatrix} 1 & 0 & 0 \\ 0 & F(a, -a, c^*, \zeta) & -\frac{ia}{c} \zeta^c F(a+c, -a+c, 1+c, \zeta) \\ 0 & -\frac{ia}{c^*} \zeta^{c^*} F(a+c^*, -a+c^*, 1+c^*, \zeta) & F(a, -a, c, \zeta) \end{pmatrix} \quad (5.18)$$

To have a unitary operation, it is necessary that Eq. (5.15) is satisfied, i.e., that the trion state is only virtually excited. Mathematically, this translates to $a = 1$. We are also only interested in the form of U after the passage of the pulse, when $\zeta = 1$. Then U becomes

$$U \simeq \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 - 1/c^* & 0 \\ 0 & 0 & 1 - 1/c \end{pmatrix} \equiv \begin{pmatrix} 1 & 0 & 0 \\ 0 & e^{-i\phi} & 0 \\ 0 & 0 & e^{i\phi} \end{pmatrix}. \quad (5.19)$$

The evolution operator is block diagonal. The 2×2 spin space evolves according to the unitary matrix:

$$U_{spin} \simeq \begin{pmatrix} 1 & 0 \\ 0 & e^{-i\phi} \end{pmatrix} = e^{-i\phi/2} \begin{pmatrix} e^{i\phi/2} & 0 \\ 0 & e^{-i\phi/2} \end{pmatrix}. \quad (5.20)$$

This phase change between the $|z\rangle$ and $|\bar{z}\rangle$ states translates to a rotation about the z axis by an angle ϕ . So, although for a true two-level system the induced phase of a 2π pulse is trivial when all the population is initially in the $|z\rangle$ state, for the three-level system it yields a non-trivial rotation about the z axis. The expression for the angle of rotation may be simplified:

$$\tan \frac{\phi}{2} = \frac{\sin \phi}{1 + \cos \phi} = \frac{\sigma}{\Delta} \implies \phi = 2 \arctan \frac{\sigma}{\Delta}. \quad (5.21)$$

A feature of this proposal as opposed to the ones reviewed above is that resonant or near resonant pulses are used. This allows for optical decay leading to lower fidelities. The way to overcome this is to use very fast pulses, which are desirable anyway. In fact in this approach, unlike the adiabatic ones, faster pulses yield higher fidelities [33].

Rotations about the x axis

Rotations about z alone do not suffice for universal quantum logic. Rotations about at least one more axis are needed⁵. In Ref. [34] we provided a way to implement rotations about the x axis, again via hyperbolic secant pulses.

It can be seen in Fig. 5.3 in the ‘ x basis’ that by choosing linearly polarized light H , we reduce the four-level system into two independent two-level systems, $\{|x\rangle, |T_x\rangle \equiv \frac{1}{\sqrt{2}}(|3/2\rangle + |-3/2\rangle)\}$ and $\{|\bar{x}\rangle, |\bar{T}_x\rangle \equiv \frac{1}{\sqrt{2}}(|3/2\rangle - |-3/2\rangle)\}$. A H linearly polarized 2π sech pulse with bandwidth β_x and frequency ω_L is used here, which acts on both transitions and which is detuned by $\Delta_1 = \omega_1 - \omega_L$ ($\Delta_2 = \omega_2 - \omega_L$) for transition $|x\rangle \leftrightarrow |T_x\rangle$ ($|\bar{x}\rangle \leftrightarrow |\bar{T}_x\rangle$). The quantities ω_1, ω_2 are the energies of the transitions $|x\rangle \leftrightarrow |T_x\rangle$ and $|\bar{x}\rangle \leftrightarrow |\bar{T}_x\rangle$ respectively. The property of the sech pulse that the pulse area of 2π is independent of the detuning is crucial: the two transitions have different detunings but the same dipole moment. So both transitions ‘see’ a 2π pulse. Then, each two-level system acquires a phase (as in Eq. 5.21) determined by its detuning. The relative phase determines the angle of rotation about the

⁵ along with conditional two-qubit control

x axis, $\phi_x = \phi_1 - \phi_2$, which is

$$\phi_x = 2 \arctan \frac{\beta_x(\Delta_1 - \Delta_2)}{\Delta_1\Delta_2 + \beta_x^2}. \quad (5.22)$$

From Eq. (5.22) a π rotation requires $\Delta_1\Delta_2 < 0$, i.e., that the common laser pulse is centered in frequency between the two transitions. This can be understood intuitively: If the laser is detuned positively (or negatively) for *both* transitions, the phases will have the same sign, so there is an upper limit to the obtainable angle of rotation. Centering the laser frequency between the two transitions gives rotation angles up to π for appropriate choice of β_x . Therefore for a π rotation there is an upper bound to the bandwidth β_x , which makes rotations about x slower than those about z .

With x and z rotations designed, all other rotations can be given as a combination of these as composite rotations. For example, rotations about the y axis are given by $R_y(\phi) = R_z^\dagger(\pi/2)R_x(\phi)R_z(\pi/2)$. Such a rotation is shown in Fig. 5.4 where the spin vector has been calculated and plotted by numerically solving the Liouville-von Neumann equations for the density matrix.

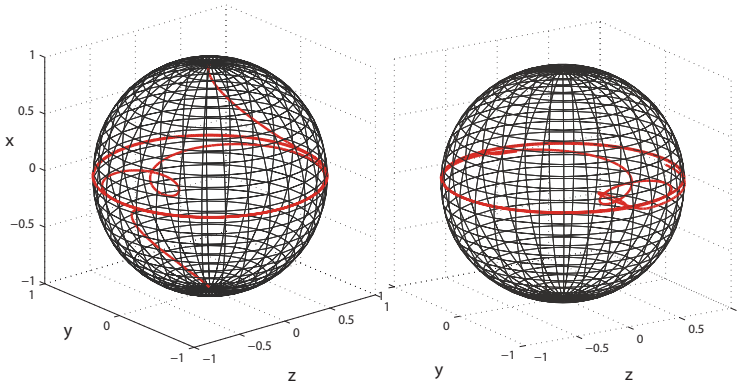


Fig. 5.4 A composite rotation of the spin about the y axis by angle π for two initial states, $|\bar{x}\rangle$ (left) and $|y\rangle$ (right). The $R_y(\pi)$ operation takes the spin vector from $-x$ to x , and leaves the y state unaffected. The plots are in the lab frame, so that the spin precession can be seen.

5.5.3.3 Use of RZ pulses and CPT for spin rotations

For QDs with larger Zeeman splittings, the pulse bandwidths discussed above can become too large for practical purposes. Then we modify our approach and use Coherent Population Trapping (CPT—see Appendix 5.6.2). CPT was first demonstrated in semiconductors for bound donors [20]. More recently it has been seen in single QDs excited with CW lasers both for electrons [23] and for holes [42].

Here we use frequency selectivity to isolate a three-level system from the four levels of Fig. 5.3, and we work in the ‘x basis’, which is the energy eigenbasis. We consider the general case where both the electron states and the trion states are Zeeman split, by ω_e and ω_h respectively. We choose narrowband pulses, $\beta \ll 2(\omega_e - \omega_h)$, where β is the bandwidth so that we can isolate a three-level system. For concreteness we spectrally focus the pulses to pick out the three-level Λ system $\{|x\rangle, |\bar{x}\rangle, |T_x\rangle\}$. The two transitions are addressed separately by polarization selectivity. The total laser field is

$$E = E_x f_x(t) e^{i\omega_x t} \hat{H} + e^{i\alpha} E_y f_y(t) e^{i\omega_y t} \hat{V} + c.c., \quad (5.23)$$

where H and V are the polarization vectors of the laser fields. For a coherently trapped state the two pulses should have the same detuning (two-photon resonance), i.e., $\omega_{x,T_x} - \omega_x = \omega_{\bar{x},T_x} - \omega_y \equiv \Delta$, and the same temporal envelope $f_x(t) = f_y(t) \equiv f(t)$. Then the new spin states, $|B\rangle$ and $|D\rangle$, are related to the basis states $\{|x\rangle, |\bar{x}\rangle\}$ through the unitary transformation

$$\mathcal{T} = \begin{pmatrix} \cos \vartheta & -e^{i\alpha} \sin \vartheta \\ e^{-i\alpha} \sin \vartheta & \cos \vartheta \end{pmatrix}. \quad (5.24)$$

Here ϑ is defined by $\tan \vartheta = E_y/E_x$, and the matrix element between $|B\rangle$ and $|T_x\rangle$ is $V_{B,T_x} = \Omega_o f(t) e^{i\Delta t}$, where $\Omega_o = \sqrt{\Omega_x^2 + \Omega_y^2}$ and Ω_x (Ω_y) is the Rabi frequency of the transition with polarization H (V). Now we choose the envelope to be $f(t) = \text{sech}(\beta t)$. We require the population to return to the spin subspace after the passage of the pulse, i.e., we require the total pulse acting on the bright state to be a 2π pulse, which gives $\Omega_o = \beta$.

We still have freedom in choosing the bandwidth β and the detuning Δ . The bandwidth is constrained by the requirement of frequency selectivity, $\beta \ll 2(\omega_e - \omega_h)$. After the passage of the pulse the bright state has picked up a phase relative to the dark state given by $\phi = 2 \arctan\left(\frac{\beta}{\Delta}\right)$. This phase is the angle by which any spin state not parallel to the B/D is rotated. The axis of rotation is that defined by B/D . By varying the laser parameters, ϑ and α , we vary the composition of the B/D basis, i.e., vary the axis of rotation. Thus we have designed an arbitrary spin rotation $R_n(\phi) = e^{-i\phi \hat{n} \cdot \sigma/2}$, where σ is the spin operator. The axis of rotation is $\hat{n} = (\cos \vartheta, \sin \vartheta \sin \alpha, \sin \vartheta \cos \alpha)$. This axis is the same as that of the adiabatic scheme described above [12] where CPT is also used. However, in that work an auxiliary lower level was required for the population to be stored during the gate. This requirement limits their scheme to physical systems where such an extra level is available. In the current approach, no auxiliary levels are needed, and the same pulse creates the trapped state and induces the phase.

5.5.3.4 Experimental demonstration

Our proposal for the rotation of spins in QDs based on hyperbolic secant pulses was recently demonstrated in an ensemble of InAs QDs for optical z rotations. In combination with the precession about the $B \parallel x$ field, they gave rotations of the spins around arbitrary directions [22]. The measurement was carried out by picosecond probe pulse ellipticity. The spins were polarized by the pump along z . Their subsequent precession about the $B \parallel x$ field can be seen in Fig. 5.5, both as a depiction of the spin on the Bloch sphere [5.5(a)] and from the actual experimental data [5.5(b)]. The traces represent the z component of the spin vector, as measured by the probe pulse. The control pulse—whose arrival time is indicated by the arrow in 5.5(b)—induces spin rotations about the z axis when the control pulse area is 2π . Since

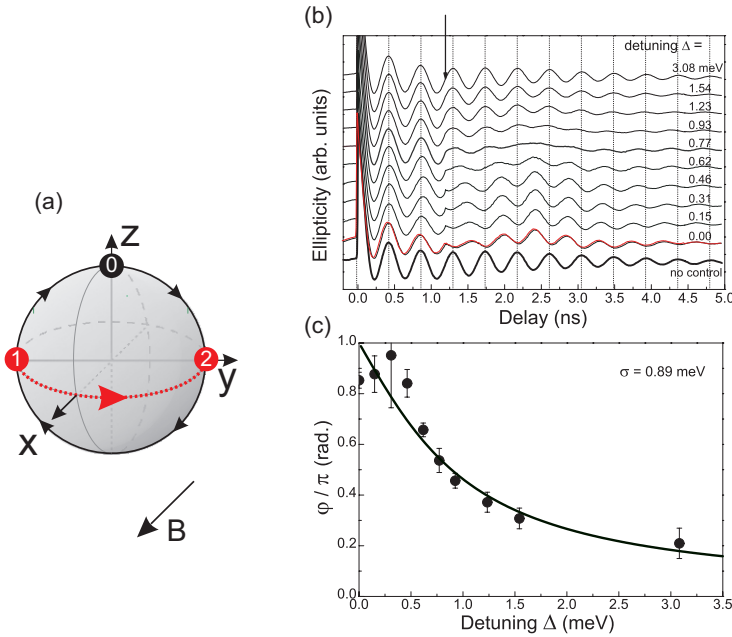


Fig. 5.5 (a) Cartoon of spin vector on Bloch sphere precessing about B field and rotated about the z axis from $-y$ to y . (b) Experimental data of the z projection of the spin for fixed pulse area 2π and varying values of the detuning. (c) Angle of spin rotation as function of detuning plotted against the theoretical expression. Figures taken from [22].

the rotations due to the control pulse are about z , the effect will be most prominent when the spins are initially in the y direction. On the other hand when the spins are in the $|\pm z\rangle$ states they are practically unaffected by the pulse. The dependence of the spin rotation angle on the detuning of the control pulse shown in 5.5(b) provides a critical test of the spin rotation proposal. The effect of a resonant pulse on the spin pointing along $-y$ is to take it to y . This is shown in the red curve in 5.5(b),

from which it can be seen that the beats have reversed phase after the action of the control pulse as compared to the reference ('no control'). For non-resonant control pulses, the angle of rotation will be $< \pi$, see Eq. (5.21), which means that the spins pointing along the y direction will acquire a component out of the yz plane, parallel to the B field. Therefore the amplitude of the oscillations of the z component will be reduced, as can be seen also from the data. We find the angle of rotation from the reduction of the amplitude. The control pulse has been fit to a sech pulse shape, from which we deduce the bandwidth to use in the expression for the rotation angle. Both the theoretical and experimental dependence of the angle of rotation on the detuning are plotted in Fig. 5.5(c). The agreement is remarkably good, especially considering that the system is inhomogeneously broadened. Thus we see that fast spin rotations have been carried out using the formulation based on sech pulses from 5.5.3.2. Further experimental details are given in [22] and in Chapter 6.

5.6 Outlook

Optical spin rotations about the z axis have been demonstrated experimentally within the last year, both for single QDs [28] and for an ensemble [22]. In both cases the rotation about axes other than z was achieved by use of the free precession of the spin(s) about the external B field. It is desirable to have rotations about these additional axes that are optically induced. The scheme described above based on hyperbolic secant pulses for x rotations should be simple for experimental demonstration, as it requires only one pulse to act on both transitions. Since the optical transition energy has to be known and narrow, it is not appropriate for ensembles where there is large inhomogeneous broadening, but it is ideal for single QDs.

The next important step in the field will be to demonstrate optical conditional control between two spins. There exist proposals for this [15, 35, 36, 37] which would suffice for a single QD molecule, but in order to scale up to a large number of spins coupling by photons in a cavity will most likely be necessary. Single or two-qubit control in a multi-qubit setting will be a challenging task, but it presents us with many theoretical and experimental research opportunities.

Appendix

5.6.1 Fidelity

It is useful to introduce the concept of fidelity, which is a measure of how close the operation we have is to the desired one. To motivate the definition, it is clear from the discussion on population transfer versus quantum operation in section 5.1

above that we need to check the effect of the operation on initial states that are pointing along inequivalent directions on the Bloch sphere. The formal definition of the fidelity is

$$f = \overline{|\langle \Psi | U^\dagger U_o | \Psi \rangle|^2}, \quad (5.25)$$

where U, U_o is the actual and the target operations respectively, and the bar denotes an average over all initial states $\{|\Psi\rangle\}$. A more useful expression can be found by parameterizing the initial states as $|\Psi\rangle = \cos \frac{\theta}{2} |0\rangle + e^{i\phi} \sin \frac{\theta}{2} |1\rangle$ and averaging over θ and ϕ (see for example [41]). The expression then for a two-level system becomes

$$f = \frac{1}{3} \sum_i |I_{ii}|^2 + \frac{1}{6} \sum_{i \neq j} (I_{ii} I_{jj}^* + I_{ij} I_{ij}^*), \quad (5.26)$$

with $I = U^\dagger U_o$.

5.6.2 Coherent Population Trapping

The phenomenon of coherent population trapping (CPT) was initially explored in the 70's by the atomic physics community. We briefly outline the main idea here as it is used in two proposals discussed above. Consider a three-level 'Λ' system, shown in Fig. 5.6. Each of the transitions is assumed addressable by a different

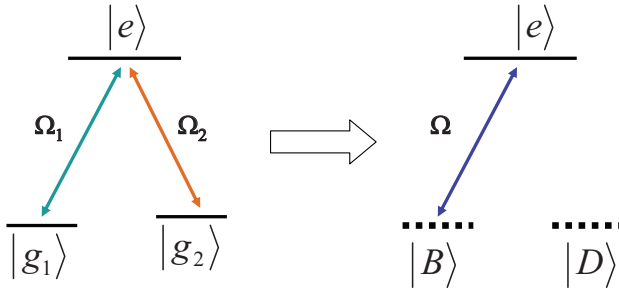


Fig. 5.6 Use of appropriate phase locked lasers reduces the three-level system to a two-level one in the interaction picture. The two new lower levels $|B\rangle$ and $|D\rangle$ are the bright and dark state respectively.

field, with Rabi frequencies Ω_1 and Ω_2 , due to their different resonance energies. The Hamiltonian is

$$H = \epsilon_1 |g_1\rangle \langle g_1| + \epsilon_2 |g_2\rangle \langle g_2| + \epsilon_e |e\rangle \langle e| + (\Omega_1 e^{i\omega_1 t} |g_1\rangle \langle e| + \Omega_2 e^{i\omega_2 t} |g_2\rangle \langle e| + h.c.), \quad (5.27)$$

where $h.c.$ stands for Hermitian conjugate. For the moment we assume that we are dealing with CW lasers, but this condition can be relaxed. In the interaction picture the Hamiltonian is:

$$\tilde{H} = \Omega_1 e^{i\Delta_1 t} |g_1\rangle\langle e| + \Omega_2 e^{i\Delta_2 t} |g_2\rangle\langle e| + h.c., \quad (5.28)$$

where $\Delta_j = \epsilon_e - \epsilon_j - \omega_j$. Under the two-photon resonance condition (i.e., when $\Delta_1 = \Delta_2 \equiv \Delta$), we have

$$\tilde{H} = e^{i\Delta t} (\Omega_1 |g_1\rangle\langle e| + \Omega_2 |g_2\rangle\langle e|) + h.c.. \quad (5.29)$$

Now we change basis to the following (the two states are degenerate in this frame due to the two-photon resonance):

$$|B\rangle = \frac{(\Omega_1 |g_1\rangle + \Omega_2 |g_2\rangle)}{\sqrt{\Omega_1^2 + \Omega_2^2}} \quad (5.30)$$

$$|D\rangle = \frac{(\Omega_1 |g_2\rangle - \Omega_2 |g_1\rangle)}{\sqrt{\Omega_1^2 + \Omega_2^2}} \quad (5.31)$$

What is special about this new basis is that if we calculate the matrix element between each of these states and the excited state $|e\rangle$ we get

$$\begin{aligned} \langle e|\tilde{H}|B\rangle &= \frac{\langle e|(\Omega_1 |e\rangle\langle g_1| + \Omega_2 |e\rangle\langle g_2| + h.c.) (\Omega_1 |g_1\rangle + \Omega_2 |g_2\rangle)}{\sqrt{\Omega_1^2 + \Omega_2^2}} \\ &= \sqrt{\Omega_1^2 + \Omega_2^2} \equiv \Omega \end{aligned} \quad (5.32)$$

and

$$\begin{aligned} \langle e|\tilde{H}|D\rangle &= \frac{\langle e|(\Omega_1 |e\rangle\langle g_1| + \Omega_2 |e\rangle\langle g_2| + h.c.) (\Omega_2 |g_1\rangle - \Omega_1 |g_2\rangle)}{\sqrt{\Omega_1^2 + \Omega_2^2}} \\ &= 0. \end{aligned} \quad (5.33)$$

That is, one state is coupled and the other one is not. Now the choice of name $|B\rangle$, $|D\rangle$ for bright and dark states becomes clear. Note that $|B\rangle$, $|D\rangle$ are bright and dark respectively for this particular laser field, meaning that the dark state has a nonzero, dipole with the excited state. Because the dipole is nonzero spontaneous emission is still allowed from $|e\rangle$ to $|D\rangle$, and therefore this technique can be used for optical pumping into the bright state. The phase between the two lasers, which has been chosen for simplicity to be zero here can also vary, and it determines the azimuthal direction of the B/D basis for a given polar direction, the latter determined by the absolute ratio of the Rabi frequencies.

This phenomenon is a simple example of destructive quantum interference: the dark state has two ways of going to the excited state which destructively interfere and the

excitation cannot take place. This effect is the basis for phenomena such as lasing without inversion and electromagnetically induced transparency. For spin rotation approaches, this technique is useful because it allows a three-level system to be reduced to a two-level system ($|B\rangle$ and $|e\rangle$).

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Chapter 6

Ensemble spin coherence of singly charged InGaAs quantum dots

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Abstract This Chapter reviews experimental and theoretical studies of electron spin coherence in singly-charged (In,Ga)As quantum dots. Ultrafast optical technique, the pump-probe Faraday rotation, is used for generation and control of the spin coherence. Despite the ensemble inhomogeneity the electron spin coherence time T_2 is measured in the mode-locking regime of synchronization of the electron spin precession about external magnetic field with the periodic laser excitation. This synchronization effect is enhanced and stabilized by electron-nuclear hyperfine interaction allowing frequency focusing of the inhomogeneous electron ensemble into the single precession mode. Ultrafast optical spin rotation of resident electrons in quantum dots to any point on the Bloch sphere is demonstrated and all-optical spin echo for these electrons is realized.

6.1 Introduction

The dynamics of carrier spins in low-dimensional semiconductor nanostructures have attracted considerable interest recently, in particular because of the potential of using them as bits in quantum information technologies. According to the criteria worked out by DiVincenzo [1] these quantum bits have to fulfill several require-

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ments, without which implementation of such technologies seems impossible. In short, these requirements are that the qubit involving two well defined spin states can be initialized, that the coherence of the qubit is maintained over times which are orders of magnitude longer than the time to perform a single operation out of a complete set of manipulations on the qubit, that an entanglement creating interaction can be established by which many qubits can be switched in parallel and that the final qubit state after the desired manipulation sequence can be read out in a single shot. Here we will present experiments which address mostly the first three, basic requirements and show that electron spins localized in self-assembled (In,Ga)As/GaAs quantum dots fulfill these conditions in a promising way.

Obviously two well defined qubit levels can be generated by application of an external field to a carrier spin, inducing the Zeeman level splitting (see Section 3, after introducing the experimental methodology in Section 2). Self-assembled (In,Ga)As/GaAs quantum dots containing an electron are well suited for these purposes, because the ground state transition obeys to a good approximation the optical selection rules of a heavy hole exciton in zincblende semiconductors, so that the spin orientation can be controlled by laser excitation (see Section 4). Further, they provide a strong confinement potential for carriers in the ground state. Due to this confinement the free orbital motion of carriers is suppressed and therefore one expects the spin relaxation mechanisms that arise from the spin-orbit coupling should be strongly reduced. These mechanisms typically provide the most efficient relaxation pathways in structures of higher dimensionality. On the other hand the carrier localization leads to an enhancement of their wave function amplitude at a particular lattice site, from which one expects an increase of the hyperfine interaction with the lattice nuclei, potentially leading to decoherence. The net impact of these influences on the electron spin coherence in quantum dots will be studied in Section 5.

The hyperfine interaction acts, however, not only destructive, but provides also an interesting way to make the initially very inhomogeneous spin precession frequency about an external magnetic field in a quantum dot ensemble much more homogeneous. This homogenization is described in Sections 6 and 7, and in effect allows to exploit the properties of a single spin properties albeit studying millions of spins. It is a specific feature of the approach taken in our studies which are not focused on single quantum dots, but on ensembles of these structures. This has the advantage of a strong interaction of the spin system with an optical light field, leading to strong spectroscopic signals and facilitating thereby short measurement times.

Irrespective of the cause of decoherence, the spin coherence time will be finite, and one has to seek for tools which allow manipulation of the spins on time scales much shorter. As typical benchmark it is often mentioned that the manipulation time has to be at least four orders of magnitude shorter than the coherence time, otherwise quantum error correction could not be implemented. Also here optical methods are very attractive because of the availability of pulsed laser systems emitting highly intense pulses with durations in the order of picoseconds or even shorter. Therefore it appears to be highly attractive to use optical tools not only for orienting spins but also for rotating them as described in Section 8.

Application of an external magnetic field B generally leads to an electron spin splitting given by $g_e\mu_B B$, where g_e is the electron g -factor and μ_B is the Bohr magneton. We will use the heterostructure growth direction z as spin quantization axis, coinciding with the optical axis in the experiment. Circularly polarized light either injects or orients electron spins along one of the spin basis states. If the magnetic field is applied along z (Faraday configuration), the electron spins are oriented along the magnetic field and are therefore stable until relaxation occurs. If the field is applied normal to the optical axis (Voigt configuration), say along the x -axis, the spins are created in a superposition of the two spin eigenstates. As a consequence the spins precess about the field with the electron Larmor frequency $\omega_e = g_e\mu_B B/\hbar$. Electron spin dynamics can be described by two phenomenological relaxation times, T_1 and T_2 , for the spin components longitudinal and transverse to external magnetic field, respectively [2]. The Faraday configuration allows one to study the longitudinal relaxation time T_1 , which describes relaxation from the upper to the lower spin split state. In finite magnetic fields, this inelastic process requires energy exchange with a surrounding bath such as the lattice phonons or nuclei, from which energy can be absorbed or into which energy can be transferred. The Voigt configuration, on the other hand, allows one to study the transverse relaxation time T_2 which is a phase relaxation time for coherent precession of the electron spin. We will refer to this time as *spin coherence time*. While inelastic scattering is one of the mechanisms that destroy the phase coherence, also elastic scattering through interaction with a bath contributes to decoherence. Therefore T_2 is typically much shorter than the longitudinal relaxation time T_1 .

Despite of intense efforts to fabricate ensembles of homogeneous quantum dots, the inhomogeneity still represents a considerable factor. This inhomogeneity is translated into the electronic band structures, and in particular into the electron g -factor. Due to variations in Larmor frequencies, the precessional motions of electron spins in different dots quickly run out of phase, a phenomenon called dephasing. Spin ensemble measurements of the transverse relaxation times are masked by dephasing, due to which the macroscopic response of the ensemble loses its coherence considerably faster than the spin coherence time T_2 of individual spins. Note, however, that coherence is retained on the individual spin level during T_2 time. The dephasing time is typically denoted as T_2^* . By introducing the inhomogeneity related relaxation time T_2^{inh} one can establish the following relation:

$$\frac{1}{T_2^*} = \frac{1}{T_2} + \frac{1}{T_2^{inh}}. \quad (6.1)$$

For an ensemble of quantum dots, each of which is loaded by a single electron, two sorts of inhomogeneities can be identified: The g -factor dispersion of the electrons dominates T_2^{inh} in strong magnetic fields, while in zero and low fields T_2^{inh} is controlled by fluctuations of the nuclear fields on the order of a few tens of mT, about which the electron spins precess as well.

In this Chapter we give an insight into the current status of coherent spin dynamics and coherent optical manipulation of electron spins in an ensemble of singly-charged (In,Ga)As/GaAs quantum dots (QD).

6.2 Experimental technique

Two modifications of the time-resolved pump-probe Faraday technique similar to those developed in Refs. [3, 4] have been used to study the coherent spin dynamics of the resident electrons in (In,Ga)As/GaAs QDs. The first technique, optically induced linear dichroism (LD), exploits linearly polarized pump pulses, which results in optical alignment of excitons in the QDs. The second technique exploits circularly polarized pump pulses for inducing circular dichroism (CD) by optical orientation of carrier spins. In both cases, the optical anisotropies induced by the pump pulse were analyzed by measuring the rotation angle of the polarization plane of a linearly polarized probe pulse of rather weak intensity [5]. The circular dichroism results from orientation of either an electron or a hole spin and, therefore, persists until the spin orientation of both carriers is destroyed. In case of linear dichroism, a coherent superposition of the $+1$ and -1 exciton states is created. This superposition is destroyed by any spin relaxation process, either electron or hole and, therefore, its decay time is limited by the fastest process. A scheme of the Faraday rotation experiment is given in Fig. 6.1(a). The sample is excited by periodic laser pulses with a duration of about 1.5 ps, emitted by a mode-locked Ti:Sapphire laser operating at a repetition rate of 75.6 MHz. This repetition rate could be reduced by a pulse picker by about three orders of magnitude. The laser beam is split into the pump and the probe pulse, which are delayed in respect to each other by a mechanical delay line. The probe pulses are linearly polarized. The rotation of the polarization plane of the transmitted probe pulses is analyzed as a function of the delay between the pump and probe pulses by means of a photodiode balanced detector. In experiments on optical spin rotation by means of the control laser pulses the second mode-locked laser has been used. It has been synchronized with the pump laser and the photon energy of the control laser can be detuned from the energy of the pump laser.

As the circular dichroism technique is the main one in our studies let's discuss it more explicitly. The quantum dot structure is excited along the z -axis by a train of circularly polarized pump pulses, which are resonant with the lowest in energy optical transition of a singly-charged quantum dot. Absorbed laser light generates an electron and a hole and excites the system from its ground state having one resident electron to the excited state with a negatively charged exciton (trion) consisting of two electrons and one hole. As a result, the resident electron spin becomes spin-oriented after trion decay. The ensemble of electron spins represents a macroscopic magnetization, which one can read out by the linearly polarized probe pulse. The rotation angle of the probe polarization plane, θ_F , is proportional to the magnetization along z -axis, see Fig. 6.1(a).

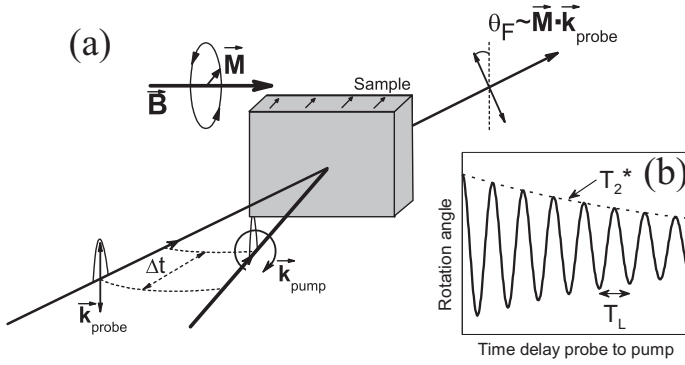


Fig. 6.1 (a) Pump-probe Faraday rotation experiment. (b) Schematic Faraday rotation signal due to electron spin precession about transverse magnetic field. $T_L = 2\pi/\omega_e$ is a period of Larmor precession

An external magnetic field B is applied normal to the structure growth direction (along the x -axis, Voigt geometry). Consequently the optically oriented electron spins precess in the $y-z$ plane about this field with a Larmor frequency given by $\omega_e = g_{e,x}\mu_B B/\hbar$, where $g_{e,x}$ is the electron g -factor along the field. As the probe maps the magnetization along z -axis, the spin precession oscillations are detected, through which one becomes also sensitive to scattering leading to reduction of this magnetization. An example of Faraday rotation trace is shown in Fig. 6.1(b). The signal contains information about the (static) spin splitting and the (dynamic) spin coherence. Its evolution as function of pump-probe time delay t can be modeled by an exponentially damped harmonic oscillator function $\exp(-t/T_2^*)\cos(\omega_e t)$. In this Chapter we report on experiments performed for ensembles of singly charged quantum dots, so that the Faraday rotation signal is strong despite the rather short signal integration times, which is achieved, however, at the expense of the ensemble inhomogeneities. An (In,Ga)As/GaAs quantum dot sample contains 20 layers of dots separated from each other by 60 nm GaAs barriers [6]. The density of dots in each layer is about 10^{10} cm^{-2} . 20 nm below each dot layer an n -doping δ -sheet with a Si-dopant density roughly equal to the dot density was placed. The as-grown InAs/GaAs sample shows ground state luminescence at a wavelength around $1.2 \mu\text{m}$ (1.05 eV) at cryogenic temperatures. After thermal annealing for 30 s up to 945°C , the ground state emission is shifted to $0.89 \mu\text{m}$ (1.4 eV) in the sensitivity range of a Si-detector, see Fig. 6.2(a). The spectral shift of the photoluminescence line in the annealed structures is explained by the fact, that during the annealing Indium ions diffuse out of the dot and are replaced by Gallium ions. As a result the binary InAs with a narrow band gap is substituted by ternary (In,Ga)As material with larger band gap energy. Additionally, due to spread of Indium ions, the QDs increase in size and their size distribution become more homogeneous. This is observed as a narrowing of the photoluminescence spectra with increasing annealing temperature,

see Fig. 6.2(a). In this chapter we will focus on the sample with 945 °C annealing temperature, which has photoluminescence line with a full width at half maximum of 15 meV. Time-resolved photoluminescence measured with a streak-camera shows a decay with a characteristic time of 360 ps, which is typical for trion and exciton radiative recombination in such quantum dots, see Fig. 6.2(b).

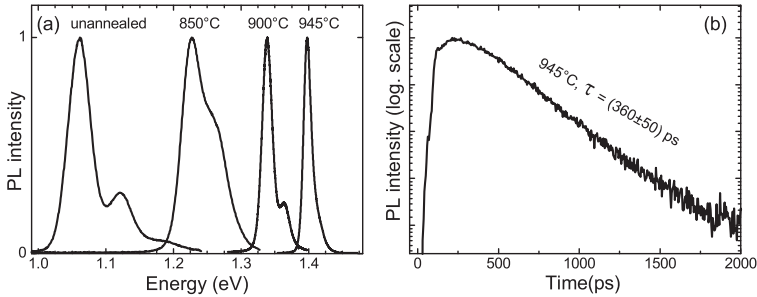


Fig. 6.2 (a) PL spectra of (In,Ga)As/GaAs QDs annealed at different temperatures. The spectra are recorded at $T = 6$ K. (b) Time-resolved photoluminescence spectra for the sample annealed at 945 °C

Faraday rotation (FR) signals measured in undoped (In,Ga)As/GaAs QDs in magnetic fields applied perpendicular to the structure growth axis, $B \parallel x$ are shown in Fig. 6.3(a). This sample is used as a reference structure to the n-doped sample which we investigate in detail in this Chapter. It is expected that in undoped QDs the generated spin coherence is provided by the photocarriers only. Therefore, its decay is controlled by the spin dephasing time and carrier recombination time, and can not exceed the last one. Indeed, the observed FR signal even in low magnetic field of 0.5 T does not extend over 1 ns and became even faster in stronger magnetic fields, where the dephasing mechanism due to g-factor inhomogeneity is due. Fast oscillations in the measured signal are caused by the electron spin beats.

Faraday rotation (FR) signals for the n-doped sample with long-lived resident electrons in QDs are shown in Fig. 6.3(b). At a magnetic field of 0.5 T the electron spin beats last beyond 2.5 ns, i.e. much longer than the trion lifetime. This allows us to assign this signal to the coherent spin precession of the resident electrons. The decay of the resident electron signal is controlled by the spin dephasing mechanisms only. This observation also confirms that in the singly charged QWs the spin coherence of the resident electrons can be generated by resonant excitation of the trion states [6]. The decay time due to dephasing strongly depends on magnetic field strength, it decreases from $T_2^* = 3$ ns at $B = 1$ T to 0.5 ns at 7 T. One can also see that in magnetic fields above 4 T additional modulation of the Faraday rotation signal appears at short delay times up to 0.5 ns. It is caused by a beating of the two close Larmor frequencies, one of the resident electron in singly charged QDs and another one of the electron in exciton in neutral dots. This modulation allows us to estimate that about 75 % of the dots are occupied by a single electron, while 25 % contain no

residual charge. Doubly occupied dots are not observed in the signals due to absence of the interaction with the resonant laser [7].

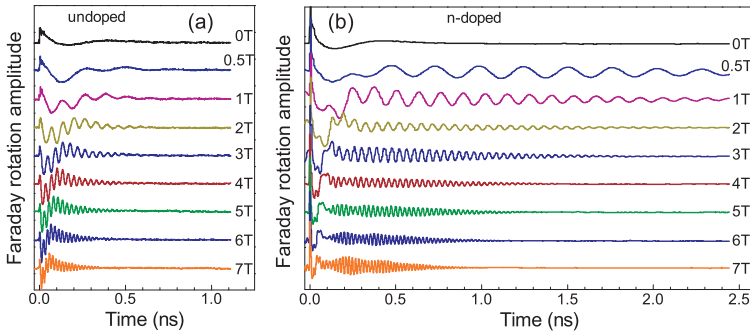


Fig. 6.3 Pump-probe Faraday rotation signals for different magnetic fields applied perpendicular to the structure growth axis (Voigt geometry). (a) Undoped reference sample with 960 °C annealing temperature, and (b) n-doped sample with 945 °C

Before we turn our attention to the underlying mechanism of the polarization of resident electrons spins, let us explore the exciton fine structure in the studied quantum dots.

6.3 Exciton fine structure

Energy splittings of QD exciton states caused by electron-hole exchange interaction (fine structure) and by magnetic field (Zeeman splitting) fall in the range of 0.01–1 meV. They are typically worked on by high-resolution spectroscopy in the spectral domain using photoluminescence or absorption spectroscopy. For that single dots have to be isolated, to reach the required spectral resolution [8]. An alternative approach is spectroscopy in the temporal domain. For measuring fine structure splitting between two levels a quantum beat spectroscopy can be used. The levels are excited coherently into a superposition state by spectrally broad laser pulses. As a result, the population of the excited levels oscillates in time with a period corresponding to the level splitting. The technique can be used for ensemble measurements [7], but one should keep in mind that the results in this case correspond to the average parameter over the large number of addressed dots.

6.3.1 Fine structure of heavy-hole exciton

In the following we briefly recapitulate the fine structure of an exciton in low-dimensional semiconductor structures.

Detailed descriptions can be found in Refs. [9, 10]. The ground state exciton is formed by an electron with spin $s = 1/2$ and a hole with angular momentum $J = 3/2$. Dimensional quantization along the structure growth axis in QDs with large width-to-height ratio causes a splitting between the light-hole and heavy-hole states. In self-assembled QDs this splitting is further enhanced by strain, and it is considerably larger than the fine structure splitting. Therefore, it is common assumption that the light holes can to a good approximation be neglected in an analysis of the ground state exciton fine structure and that only the heavy holes with $J_z = \pm 3/2$ need to be considered. The exciton states formed with heavy-holes have angular momentum projections $M = J_z + s_z = \pm 1, \pm 2$. The origin of the angular momentum coupling is the electron-hole exchange interaction. The selection rules for left/right circularly polarized light allow optical transitions to exciton states with $M = \pm 1$ (the so-called "bright" states, $|\pm 1\rangle$). Optical transitions to states with $M = \pm 2$ ("dark" states, $|\pm 2\rangle$) are forbidden in dipole approximation.

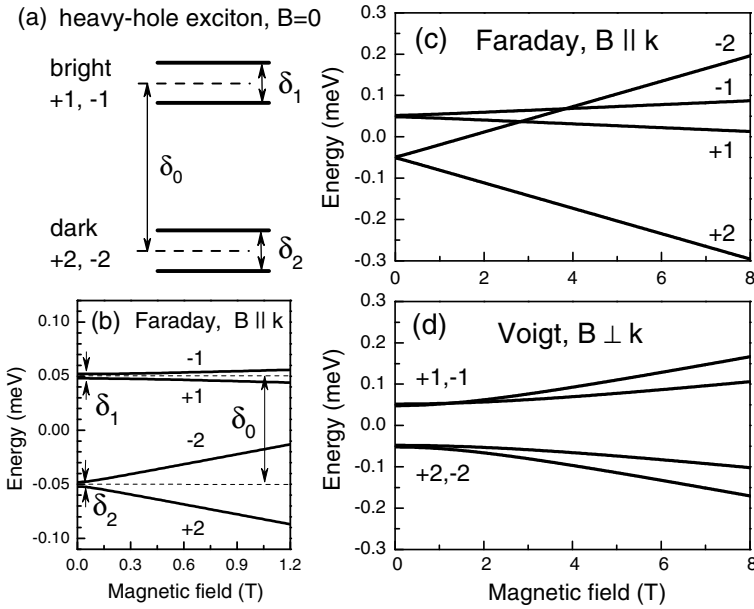


Fig. 6.4 Fine structure of a heavy-hole exciton in magnetic fields calculated for (In,Ga)As/GaAs QD with parameters given in Section 6.3.5. (a) Schema in the absence of external magnetic field. Splitting in longitudinal magnetic field (Faraday geometry) is given in (b, c). (d) Splitting in transverse magnetic field (Voigt geometry).

Phenomenologically the fine structure of an exciton can be analyzed by an effective spin-Hamiltonian similar to those used in Refs. [9, 11], in which the hole is reduced to an effective spin $1/2$ particle:

$$\mathcal{H}_X = \sum_{i=x,y,z} [\mu_B (g_{e,i} s_i + g_{h,i} \tilde{S}_{h,i}) B_i - c_i s_i \tilde{S}_{h,i}]. \quad (6.2)$$

Here $\tilde{S}_h$ is the effective hole spin operator with components $\tilde{S}_h = \pm 1/2$ corresponding to $J_z = \pm 3/2$. $g_{e,i}$ and $g_{h,i}$ are the electron and hole g -factor tensor components. Here the '+' sign of g -factor is defined such that for $g > 0$, the spin-down state of a particular excitation (i.e. a $-1/2$ electron, a -1 exciton, or a $-3/2$ hole) is the lowest in energy, i.e. is predominantly populated at low temperatures [7]. In absence of a magnetic field the fine structure is given by the electron-hole exchange determined by the interaction parameters c_i . In accordance with Refs. [9, 11] the relevant exchange splittings are

$$\delta_0 = c_z/4, \quad \delta_1 = -(c_x + c_y)/4, \quad \delta_2 = -(c_x - c_y)/4. \quad (6.3)$$

δ_0 is the component in the exchange interaction, which gives the splitting between bright and dark excitons in the isotropic case, while δ_1 and δ_2 are the anisotropic components giving the splitting of the bright as well as dark excitons, respectively, see Fig. 6.4(a). Due to the anisotropy δ_1 , the optically active eigenstates Ψ_1 and Ψ_2 are symmetric and antisymmetric combinations of the circularly polarized states $|\pm 1\rangle$. δ_2 does the same for the dark excitons. The transitions to the Ψ_1 and Ψ_2 states correspond to linearly polarized oscillators which are orthogonal to each other. By choosing pump pulses with a linear polarization different from the two exciton dipoles and with a duration so short that the corresponding spectral width covers δ_1 , both states can be excited into a superposition, for which we expect in the linear dichroism signal oscillations with a frequency $\omega = \delta_1/\hbar$. The corresponding trace recorded at zero magnetic field is shown in Fig. 6.5(a). Damping of this signal is caused by variation $\Delta\omega$ in the inhomogeneously broadened QD ensemble. By fitting the data $\delta_1 = (4 \pm 4) \mu\text{eV}$ has been evaluated.

6.3.2 Linear dichroism in longitudinal magnetic field

The Zeeman interaction in a longitudinal magnetic field ($B \parallel z$) is described by the first summand of Eq. (6.2) for which the angular momentum levels $|\pm 1\rangle$ and $|\pm 2\rangle$ are the eigenstates. The magnetic field induces a Zeeman splitting between the two bright as well as the two dark excitons (Fig. 6.4(b)). The energies of the optically active states $|\pm 1\rangle$ are given by:

$$E_{1,2} = \frac{\delta_0}{2} \pm \frac{1}{2} \sqrt{(g_{h\parallel} - g_{e\parallel})^2 \mu_B^2 B^2 + \delta_1^2}, \quad (6.4)$$

where $g_{e\parallel}$ and $g_{h\parallel}$ are the longitudinal components of the electron and hole g -factors along the z -direction. Coherent excitation results in quantum beats with frequency $\omega = (E_1 - E_2)/\hbar$ as shown in Fig. 6.5(a) [12, 13]. The exciton g -factor, $g_{X\parallel} = g_{h\parallel} - g_{e\parallel}$, can be extracted from the dependence of beat frequencies on magnetic field (Fig. 6.5(b)): $|g_{X\parallel}| = |g_{h\parallel} - g_{e\parallel}| = 0.16 \pm 0.11$. The experiments in tilted magnetic fields allow us to measure separately the longitudinal g -factors for electrons and holes: $|g_{e\parallel}| = 0.61$ and $|g_{h\parallel}| = 0.45$.

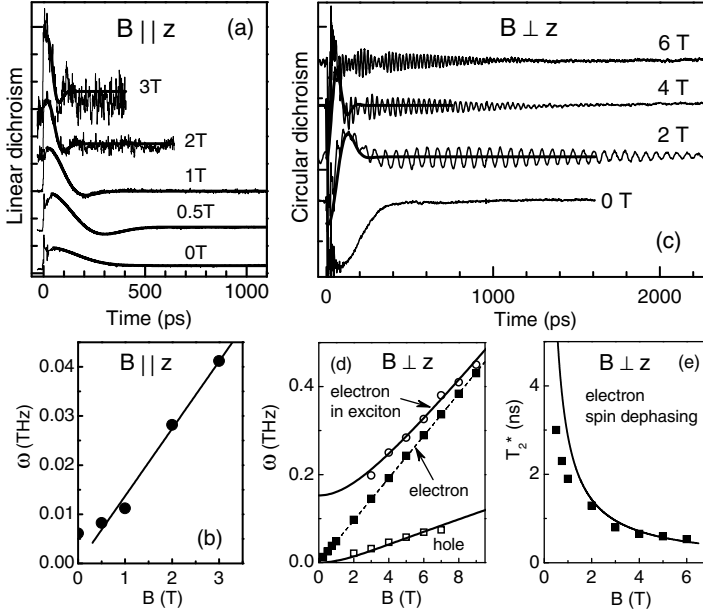


Fig. 6.5 (a) Linear dichroism signals in longitudinal magnetic fields (Faraday geometry). Thick lines are fits to the data by exponentially damped harmonics. (b) Field dependence of the precession frequency extracted from the fits. Line is a B -linear fit. (c) Circular dichroism traces in transverse magnetic fields (Voigt geometry). Lines at 2 and 4 T give fits to the initial part of the traces by exponentially damped harmonic functions. (d) Spin beat frequencies vs magnetic field for the long-lasting oscillations (solid squares) as well as for the initial part oscillations (open circles and open squares). Dashed line is a B -linear fit. Solid lines are fits of the exciton fine structure. (e) Spin dephasing time T_2^* of the long-lasting oscillations as function of magnetic field. The line gives a $1/B$ fit. $T = 2$ K.

6.3.3 Circular dichroism in transverse magnetic field

For completeness, circular dichroism signals are presented in Fig. 6.5(c). Quantum beats with at least two different frequencies are clearly observed, resulting in a modulation of the signal at short delay times. The long-lived component of the circular dichroism signal has a frequency $\omega_e = g_{e\perp} \mu_B B / \hbar$, where $g_{e\perp}$ is the electron g -factor in the QD plane. From a B -linear fit given by a dashed line in Fig. 6.5(d) $|g_{e\perp}| = 0.54$ is obtained. The dephasing time of spin ensemble T_2^* is contributed by the individual spin coherence time T_2 and the inhomogeneous spin relaxation time $T_2^{inh} = \hbar / \Delta g_e \mu_B B$. T_2^* shows a $1/B$ dependence (Fig. 6.5(e)) allowing with the use of Eq. (6.1) obtain the spread of the electron g -factor $|\Delta g_{e\perp}| = 0.005$.

A Fourier analysis of the circular dichroism signals during the first 0.5 ns shows that the three frequencies are contributed. One of it coincides with the electron frequency seen at longer delays. Two others shown by open symbols in Fig. 6.5(d) belongs to the exciton fine structure.

In the absence of anisotropic exchange, a transverse magnetic field mixes each of the "bright" $|\pm 1\rangle$ states with both "dark" states $|\pm 2\rangle$. The presence of anisotropic exchange results in mixing of all four states (Fig. 6.4(d)). In general quantum beats with six frequencies are expected for circular dichroism signals. Accounting that $\delta_0 \gg \delta_1 > \delta_2$ leaves four different frequencies [14]. This number is further reduced to two if the transverse heavy-hole g -factor, $g_{h\perp}$, is nonzero, but much smaller than $g_{e\perp}$ and than $\delta_0/\mu_B B$ [13, 15]. That is expected for QDs due to the large diameter-to-height ratio. The exciton fine structure splitting received from Eq. (6.2) is [7]:

$$\omega_{1,2} \approx \frac{\sqrt{\delta_0^2 + (g_{e\perp}\mu_B B)^2}}{\hbar}, \quad \omega_{3,4} \approx \frac{g_{e\perp}g_{h\perp}\mu_B^2 B^2}{\hbar\sqrt{\delta_0^2 + (g_{e\perp}\mu_B B)^2}}. \quad (6.5)$$

In low magnetic fields $\omega_{1,2}$ increase like B^2 starting from $\delta_0/\hbar$. In high fields $\delta_0 \ll g_{e\perp}\mu_B B$ so that $\omega_{1,2}$ are given by the electron Zeeman splitting (open circles in Fig. 6.5(d)). $\omega_{3,4}$ tend to zero for low fields, while in high fields they are determined by the hole spin-splitting: $\omega_{3,4} \approx g_{h\perp}\mu_B B/\hbar$ (open squares). By fitting these experimental data (solid lines) with the non-approximate forms given in Ref. [7] one obtains: $\delta_0 = 0.10 \pm 0.01$ meV and $|g_{h\perp}| = 0.15$.

6.3.4 Spectral dependence of the electron g -factor

The energy dispersion of the electron g -factor within the dot ensemble has been measured by varying the excitation energy across the emission band. Figure 6.6(a) shows that from the low to the high energy side the g -factor decreases from 0.57 to 0.53. This variation can be understood if one makes the assumption that the main effect of the confinement is an increase of the bandgap E_g . The deviation of g_e from the free electron g -factor $g_0 = 2$, determined from $\mathbf{k} \cdot \mathbf{p}$ calculations, is given by [16, 17]:

$$g_e = g_0 - \frac{4m_0 P^2}{3\hbar^2} \frac{\Delta}{E_g(E_g + \Delta)}. \quad (6.6)$$

Here m_0 is the free electron mass, P is the matrix element describing the coupling between valence and conduction band, and Δ is the spin-orbit splitting of the valence band. The decrease of the g -factor modulus with increasing emission energy could be then only explained if the g -factor has a negative sign. This argument is supported by measurements of the dynamic nuclear polarization [7], similar to those described in [18]. This allows us to determine also signs for the exciton and hole g -factors.

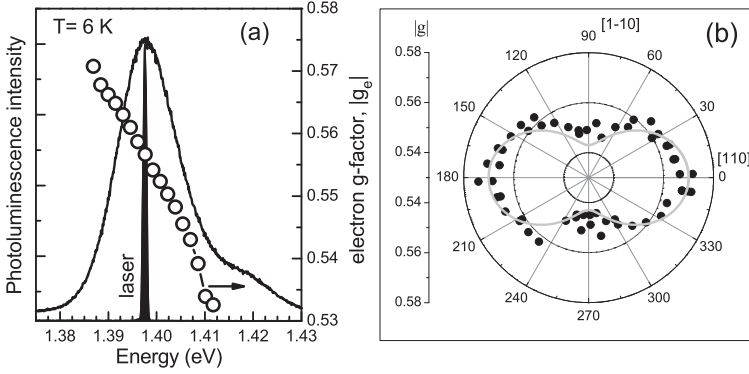


Fig. 6.6 (a) Photoluminescence spectrum of an (In,Ga)As/GaAs quantum dot sample. The filled trace gives the spectrum of the excitation laser used in the Faraday rotation experiments, which could be tuned across the inhomogeneously broadened emission band. The symbols give the electron in-plane g -factor along the $[1\bar{1}0]$ direction across this band. (b) In-plane angular dependence of the electron g -factor. Line is a fit to the data using Eq.(6.7). $B = 5$ T. Angle zero corresponds to a field orientation along the x -direction which is defined by the $[110]$ crystal axis [7].

6.3.5 Anisotropy of electron g -factor in quantum dot plane

By varying the field orientation in the quantum dot plane one can determine the in-plane anisotropy of the electron g -factor. For an arbitrary direction, characterized by the angle α relative to the x -axis, the electron g -factor is

$$|g_{e\perp}(\alpha)| = \sqrt{g_{e,x}^2 \cos^2 \alpha + g_{e,y}^2 \sin^2 \alpha}, \quad (6.7)$$

where $g_{e,x}$ and $g_{e,y}$ are the g -factors along the x and y -axes, $[110]$ and $[1\bar{1}0]$, respectively. The circular dichroism signal has been measured as a function of α . Figure 6.6(b) shows the resulting angular dependence of the electron g -factor. One finds from fit by Eq.(6.7) $|g_{e,x}| = 0.57$ and $|g_{e,y}| = 0.54$. The origin of the in-plane anisotropy is related to the shape anisotropy of quantum dots and is well described by a tight-binding calculations [19].

To conclude on the results presented in Section 6.3, a pump-probe Faraday rotation technique in an external magnetic field offers a detailed insight into the exciton fine structure parameters. For the studied self-assembled (In,Ga)As/GaAs QDs the following values of the longitudinal and transversal g -factors of electron, hole and exciton have been obtained: $g_{e\parallel} = -0.61$, $g_{e\perp} = -0.543 \pm 0.005$, $g_{h\parallel} = -0.45$, $|g_{h\perp}| = 0.15 \pm 0.05$, and $g_{X\parallel} = g_{h\parallel} - g_{e\parallel} = 0.16 \pm 0.11$. Further, for the electron g -factor we have found a remarkable in-plane anisotropy. Also the exchange constants for the QD excitons have been measured: $\delta_0 = 0.10 \pm 0.01$ meV and $\delta_1 \sim 0.004 \pm 0.004$ meV.

6.4 Generation of spin coherence

We turn now to the generation of spin coherence for resident electrons in singly charged quantum dots under resonant excitation into a trion. It is documented by the experimental results in Fig. 6.3(b) that the electron spin coherence can be generated optically. Deeper insight into the responsible mechanism is provided by the pump density dependence of the generation efficiency. Figure 6.7(a) shows Faraday rotation amplitudes for different pump powers, or the laser pulse area Θ defined as $\Theta = 2 \int [\mathbf{d} \cdot \mathbf{E}(t)] dt / \hbar$ in dimensionless units. $\mathbf{d}$ is the dipole matrix element for the transition from the valence to the conduction band. $\mathbf{E}(t)$ is the electric field amplitude of the laser pulse. For pulses of constant duration, Θ is proportional to the square root of excitation power.

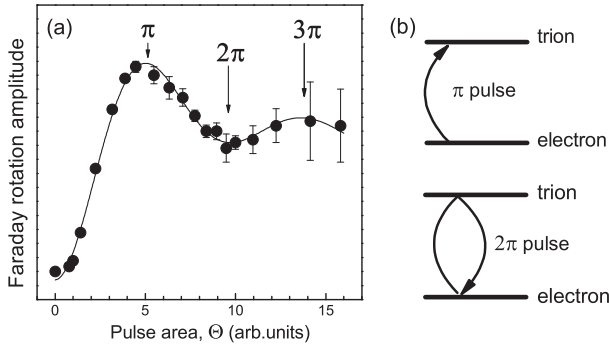


Fig. 6.7 Pump power dependence of the spin coherence generation in (In,Ga)As/GaAs quantum dots. (a) Faraday rotation amplitude versus laser pulse area Θ . The line is a guide to the eye [6]. (b) The scheme illustrates the generation process for π and 2π -pulse

The Faraday rotation amplitude shows a non-monotonic behavior with increasing pulse area. First, it rises to reach a maximum, then drops to about 60 %. Thereafter it shows another strongly damped oscillation. This behavior is similar to the one known from Rabi-oscillations of a Bloch vector, whose z -component describes the electron-hole population [20, 21]. The laser pulse drives coherently this population, leading to its oscillations as a function of the pulse area Θ . In this case the ground state is an empty dot, and the excited state represents a dot with a photogenerated electron-hole pair. For the case of a singly charged dot, the ground state is a dot with an electron and the excited state is a dot with a trion, i.e. a dot containing two electrons and one hole. A laser pulse with $\Theta = \pi$ drives the system from the ground to the excited state, which corresponds to maximal generation efficiency, see Fig. 6.7(b). Further increase of the laser power does not increase the generated spin polarization. The reason is that the very same pulse with $\Theta > \pi$ starts to drive the system back to the ground state. For $\Theta = 2\pi$ the system returns to the ground state and no spin polarization is generated. Thus the Faraday rotation amplitude achieves a maximum for a π -pulse, and a minimum for a 2π -pulse, see Fig. 6.7(a).

The damping of the oscillations most likely is due to ensemble inhomogeneities of quantum dot properties such as the dipole moment d [22].

With these observations at hand one can understand the origin of the spin coherence generated in quantum dots. We discuss first neutral dots. Resonant optical pulses with σ^- polarization create a superposition state of vacuum and exciton:

$$\cos\left(\frac{\Theta}{2}\right) |0\rangle - i \sin\left(\frac{\Theta}{2}\right) |\uparrow\downarrow\rangle, \quad (6.8)$$

where $|0\rangle$ describes the ground state of the semiconductor QD. The hole spin orientations $J_z = \pm 3/2$ are symbolized by the arrows $\uparrow$ and $\downarrow$, respectively. The electron and hole spins become reversed for σ^+ polarized excitation. The exciton component precesses in magnetic field for a time, which cannot last longer than the exciton lifetime. In an ensemble, the precession can be seen until the coherence is destroyed by spin scattering of either electron or hole. The strength of the contribution to the ensemble Faraday rotation signal is given by the square of the exciton amplitude $\sin^2(\Theta/2)$.

Let us turn now to singly charged quantum dots, for which the resonant excitation can lead to the excitation of trions. We assume that the deexcited quantum dot state is given by an electron with arbitrary spin orientation:

$$\alpha |\uparrow\rangle + \beta |\downarrow\rangle, \quad (6.9)$$

with $|\alpha|^2 + |\beta|^2 = 1$. A σ^- polarized laser pulse would create an exciton with spin configuration $|\uparrow\downarrow\rangle$. This is, however, restricted by the Pauli-principle, due to which the optically excited electron must have a spin orientation opposite to the resident one in order to form a trion singlet state $|\uparrow\downarrow\downarrow\rangle$, which consists of two spin singlet electrons and a hole in state $|\downarrow\rangle$. Therefore, the pulse excites only the second component of the initial electron state, i.e. $\beta |\downarrow\rangle$. As a consequence, a coherent superposition state of an electron and a trion is created:

$$\alpha |\uparrow\rangle + \beta \cos\left(\frac{\Theta}{2}\right) |\downarrow\rangle - i\beta \sin\left(\frac{\Theta}{2}\right) |\downarrow\uparrow\downarrow\rangle. \quad (6.10)$$

We assume that decoherence does not occur during the excitation process, i.e., the pulse length is much shorter than the trion radiative decay and the carrier spin relaxation times. One can see from Eq. (6.10) that the electron-hole population should oscillate with pulse area Θ . The excitation is most efficient for $\Theta = \pi$, which gives the superposition state:

$$\alpha |\uparrow\rangle - i\beta |\downarrow\uparrow\downarrow\rangle. \quad (6.11)$$

After some time the electron-hole pair will relax, leaving only the electron part in the quantum dot. This occurs on the time scale of the trion radiative recombination. At zero magnetic field and in the absence of hole spin relaxation within the trion, the system will return to its initial state described by Eq. (6.9). Therefore, no electron

spin coherence will be generated. However, spin coherence will be generated, if the hole spin relaxation takes place before the trion recombination. In an external magnetic field in the Voigt geometry, the hole relaxation is no more the crucial factor for spin coherence generation. The reason is that the electron part of Eq. (6.11), i.e. $\alpha |\uparrow\rangle$ will precess around the magnetic field, while the singlet state of trion does not precess. Therefore, full compensation of the induced spin polarization after trion recombination is impossible and spin coherence is induced. Modeling of the electron and trion spin dynamics under pulsed resonant excitation in terms of the electron and trion spin vectors, S and J , can be described by the vector equations [23, 6]:

$$\begin{aligned}\frac{dJ}{dt} &= [\omega_h \times J] - \frac{J}{\tau_s^h} - \frac{J}{\tau_0^T} \\ \frac{dS}{dt} &= [(\omega_e + \omega_N) \times S] - \frac{S}{\tau_s^e} + \frac{(Jz)z}{\tau_0^T},\end{aligned}\quad (6.12)$$

where $\omega_{e,h}$ are electron and hole spin precession frequencies, and $\omega_N = g_e \mu_B B_N / \hbar$ is the electron precession frequency in an effective nuclear field, B_N . Both vectors: $\omega_{e,h} \perp z$, where z is the unit vector along the z -axis. τ_s^e and τ_s^h are the electron and hole spin relaxation times, and τ_0^T is the spontaneous recombination time of the trion. At low temperatures, τ_s^e is at least on the order of μs and is mainly determined by fluctuations of the nuclear field in a single QD [26, 27, 24, 25]. This time scale is irrelevant to our problem, therefore we can neglect the term S/τ_s^e in Eq. (6.12). The spin relaxation time of the hole in the trion, τ_s^h , is caused by phonon assisted process and at low temperatures may be as long as τ_s^e [28, 29].

The first part of Eq. (6.12) gives the following time dependence for the trion polarization:

$$J_z(t) = J_z(0) \exp(-i\omega_h t - \gamma_T t), \quad (6.13)$$

where $\gamma_T = 1/\tau_0^T + 1/\tau_s^h$ is the total spin decoherence rate of the trion. Substituting $J_z(t)$ into the second part of Eq. (6.12) one can obtain the following equation for the electron spin polarization:

$$\frac{dS}{dt} = [(\omega_e + \omega_N) \times S] + \frac{z}{\tau_0^T} J_z(0) \cos(\omega_h t) \exp(-\gamma_T t). \quad (6.14)$$

Equation (6.14) can be solved for the complex spin components $S_{\pm} = S_y \pm iS_z$ representing rotation of the spin around the magnetic field oriented along the x -axis. The important characteristic of the electron spin precession is the imaginary part of $S_+(t)$, which describes the electron spin polarization along the z -axis, that is measured by Faraday rotation. After trion recombination ($t \gg \tau_0^T$), the amplitude of the long-lived electron spin polarization excited by a $(2n+1)\pi$ -pulse is given by

$$S_z(t) = \text{Re} \left\{ \left[S_z(0) + \frac{J_z(0)}{2\tau_0^T} \left(\frac{1}{\gamma_T + i(\omega + \omega_h)} + \frac{1}{\gamma_T + i(\omega - \omega_h)} \right) \right] e^{i\omega t} \right\}, \quad (6.15)$$

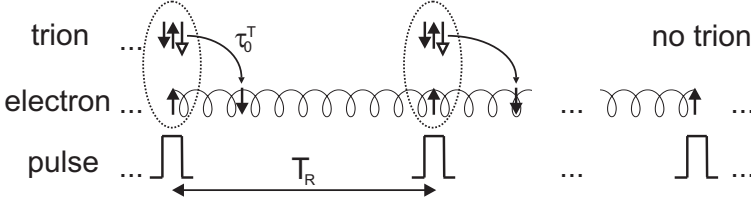


Fig. 6.8 Sketch illustrating the electron spin polarization by the periodic excitation in magnetic field. By initial excitation, two components are present, trion (β) and dark electron (α). If electron component precess with an integer number of $2\pi/\omega$ it stays dark for all following pulses as long as it keeps coherence. The trion component decays at different times, and the released electron continue to precess until next pulse, where again some part of its polarization is bounded to the trion. This process continues until 100% of electron polarization is achieved ($\alpha = 1$) and no trions are created any more ($\beta = 0$).

where $S_z(0)$ and $J_z(0)$ are the electron and trion spin polarizations created by the pulse, and $\omega = \omega_e + \omega_N$ is the electron precession frequency in the magnetic field resulting from the external field and the effective nuclear field. If the radiative relaxation is fast $\tau_0^T \ll \tau_s^T, \omega_{e,h}^{-1}$, the induced spin polarization $S_z(t)$ is nullified on average by trion relaxation, as $S_z(0) = -J_z(0)$. This corresponds to the situation at zero magnetic field. In contrast, if the spin precession is fast, $\omega_{e,h} \gg (\tau_0^T)^{-1}$, the electron spin polarization is maintained after trion decay [23, 30]. It is the case for the studied (In,Ga)As/GaAs dots.

Additionally, by subsequent application of laser pulses the spin polarization can be maximized. If the pump pulse separation is $2\pi N/\omega$ (N is an integer), the created electron spin polarization by the previous pump pulse will not be affected by the next pump pulse due to Pauli blocking. In the time between the pulses, the electrons bound to the trions are not affected by the magnetic field, so they keep their polarization. After trion recombination, which happens at different times, the released electrons also start to precess around the magnetic field, and by the next pump pulse some part of its polarization goes into synchronized electron polarization and smaller part of electron polarization is bounded to the trion. Therefore, in the case of π -pulses a dozen of them is sufficient to create a 99 % spin polarization [23], see Fig. 6.8. This requires of course that the electron spin precession frequency in external and nuclear fields fulfills the condition $\omega \equiv \omega_e + \omega_N = 2\pi N/T_R$, where T_R is the repetition period of the laser. More pulses are needed if the pulse power deviates from that of a π -pulse [31].

6.5 Mode-locking of spin coherence

As already mentioned in the Section 6.2, the ensemble dephasing does not lead to a destruction of the spin coherence in individual quantum dots, but masks it due to

the rapid accumulation of phase differences for electron spin precession in different dots.

To address this point, we look again at Faraday rotation traces, especially for negative delays shown in Fig. 6.9(a). Long-lived electron spin beats are seen at positive delays, as discussed before. Surprisingly, also for negative delays strong spin beats with frequencies similar to those of the electron precession are observed. The amplitude of these beats increases when approaching zero delay $t = 0$. Note, that spin beats at negative delay have been reported for experimental situations in which the dephasing time exceeds the time interval between the pump pulses: $T_2^* \geq T_R$ [32]. This is clearly not the case here because the Faraday rotation signal has fully vanished after 1.5 ns at $B = 6$ T (Fig. 6.9(a)), so that the dephasing is much faster than the pulse repetition period. The frequency and rise time of the signal at negative delays are the same as at positive delays, indicating that the negative delay signal is also given by the electron spin precession.

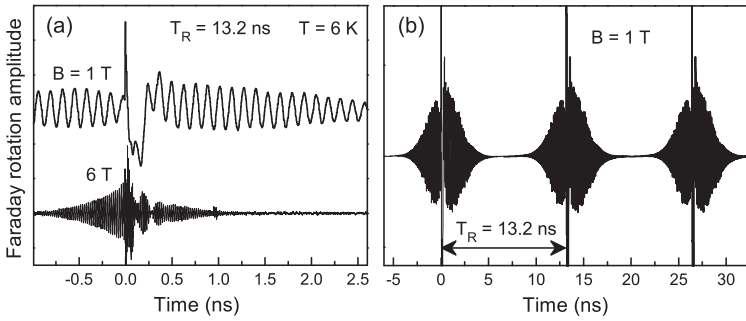


Fig. 6.9 (a) Pump-probe Faraday rotation signals at different magnetic fields in singly charged (In,Ga)As/GaAs quantum dots. The pump power density is 60 W/cm^2 , the probe density is 20 W/cm^2 [31]. (b) Faraday rotation signal recorded for a longer delay range in which three pump pulses were located

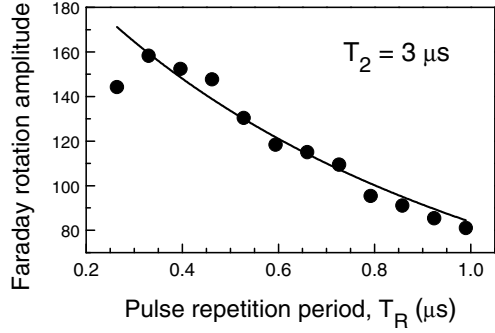
Figure 6.9(b) shows the signal when scanning the delay over a larger time interval, in which three pump pulses, separated by 5.2 ns from each other, are located. At each pump pulse arrival electron spin coherence is created, which is dephased after a few ns. Before the next pump arrival electron coherent signal reappears. This negative delay precession can occur only if the coherence of the electron spin in each dot prevails for much longer times than T_R , i.e. if $T_2 \gg T_R$.

6.5.1 Spin coherence time of an individual electron

Independent of the origin of the coherent signal at negative delays, its observation opens a pathway towards measuring the spin coherence time T_2 : When increasing the pump pulse separation until it becomes comparable with T_2 , the amplitude of the

negative delay signal should decrease. Corresponding data are given in Fig. 6.10. The Faraday rotation amplitude detected at a fixed negative delay shortly before the next pump arrival is shown there as function of T_R . T_R is increased from 5.2 up to 990 ns. A decrease of the amplitude is seen, demonstrating that T_R becomes comparable to T_2 . The result of model calculations shown by the line allow us to evaluate the electron spin coherence time $T_2 = 3.0 \pm 0.3 \mu\text{s}$, which is by four orders of magnitude longer than the ensemble dephasing time $T_2^* = 0.4 \text{ ns}$ at $B = 6 \text{ T}$.

Fig. 6.10 Faraday rotation amplitude at negative delay as function of the time interval between subsequent pump pulses measured at $B = 6 \text{ T}$ and $T = 6 \text{ K}$. The line shows calculations with a single fit parameter $T_2 = 3 \mu\text{s}$ [31]



6.5.2 Mechanism of spin synchronization

As already explained in the Section 6.4 if the pulse period, T_R , is equal to an integer number N times the electron spin precession period, $2\pi/\omega$, the action of such π -pulses leads to almost complete electron spin alignment along the light propagation direction z [23]. In general, the degree of synchronization for π -pulses is given by $P_\pi = \exp(-T_R/T_2)/[2 - \exp(-T_R/T_2)]$. In our case it reaches its largest value $P_\pi = 1$, corresponding to 100% synchronization, because for excitation with high repetition rate such as 75.6 MHz the repetition period $T_R \ll T_2$ so that $\exp(-T_R/T_2) \approx 1$.

We remind that in an ensemble of quantum dots, the electrons do not precess with the same frequency, but have a frequency distribution with a broadening γ . The latter is determined by the electron g -factor dispersion and the spectral width of the pump laser (Fig. 6.6(a)). The ensemble contains quantum dots whose electron spin precession frequencies fulfill a synchronization relation with the laser, which we term phase synchronization condition (PSC) :

$$\omega = 2\pi N/T_R \equiv N\omega_R. \quad (6.16)$$

Here ω_R is the repetition frequency of the pump pulses. The PSC modes selected from the continuous distribution of electron precession frequencies are shown by the dashed lines in Fig. 6.11. Since the electron spin precession frequency is typically

much bigger than the laser repetition rate, we have $N \gg 1$ for not too small magnetic fields. Since in addition the spread of precession frequencies is also much larger than the laser repetition rate, multiple subsets within the optically excited quantum dot ensemble satisfy Eq. (6.16) for different N . This is illustrated schematically by Fig. 6.11. In the left side three precession modes satisfying the PSC Eq. (6.16) with $N = 6, 7, 8$ are given. The spin precession with a frequency different from the PSC is shown in the right side of the figure. Two important conclusions can be drawn from this:

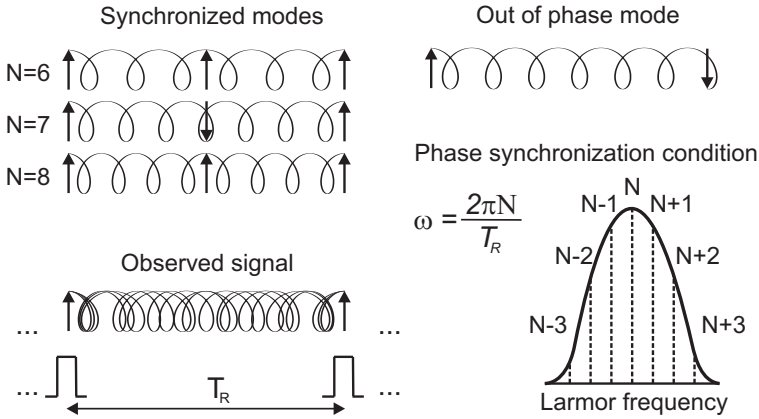


Fig. 6.11 Scheme of the phase synchronization condition for the electron spin coherence with a periodic train of laser pulses. On the left modes satisfying the PSC of Eq. (6.16) are shown. On the right a non-PSC dot is given. The arrows show the spin orientation of a resident electron in a quantum dot. Bottom right the thick solid line shows the distribution of electron precession frequencies in the dot ensemble caused by the dispersion of the electron g -factor. The PSC modes selected from this distribution are shown by the dashed lines

First, for the PSC dots spin synchronization will be accumulated until it reaches its maximum value, see discussion above for the single dot synchronization. The reason is that at the moment of the pump pulse arrival the spin coherence generated by the previous pulse has the same orientation as the one which the subsequent pulse induces. In other words the contributions of all pulses in the train are constructive. In contrast, the contributions have arbitrary orientations in the non-PSC dots (right panel of Fig. 6.11) and for these dots the degree of spin synchronization will be always far from saturation. Practically, this means that a PSC dot gives a contribution to the Faraday rotation signal than a non-PSC dot.

Second, in the PSC dots (left panels) the electron spins indicated by the arrows have the same phase at the moments of pump pulse arrival, but they have different orientation between the pulses. As a result, the signal from the dot subsets satisfying the PSC dephases shortly after each pump pulse. However it is revived before the next pulse leading to the characteristic signal shown in Fig. 6.9(b). To be more specific, the spins in each PSC subset precess between the pump pulses with fre-

quency $N\omega_R$, starting with an initial phase which is the same for all subsets. Their contribution to the spin polarization of the ensemble at a time t after the pulse is given by $-0.5\cos(N\omega_R t)$. The sum of oscillating terms from all synchronized subsets leads to a constructive interference of their contributions to the Faraday rotation signal around the times of pump pulse arrival (Fig. 6.11 bottom left). The rest of the quantum dots do not contribute to the average electron spin polarization $\bar{S}_z(t)$ at times $t \gg T_2^*$, due to dephasing. The synchronized spins therefore move on a background of dephased electrons, which however could precess individually during the spin coherence time, if they would not be repolarized at the every pump pulse arrival. The number of synchronized PSC subsets can be estimated by: $M \sim \gamma/\omega_R$. It increases linearly with magnetic field and T_R . We will discuss this in more detail in Section 6.7.

The π -pulse excitation is not critical for the electron spin phase synchronization by the pulse train. Any resonant pulse train of arbitrary intensity creates a coherent superposition of trion and electron in a quantum dot, leading to a long-lived coherence of the resident electron spins, because the coherence is not affected by the radiative decay of the trion component. Each pulse of σ^- polarized light changes the electron spin projection along the light propagation direction by $\Delta S_z = -(1 - 2|S_z(t \rightarrow t_n)|)W/2$, where $t_n = nT_R$ is the time of the n -th pulse arrival, and $W = \sin^2(\Theta/2)$ [6, 33]. Consequently, a train of such pulses orients the electron spin along the light propagation direction, and it also increases the degree of electron spin synchronization P . Application of $\Theta = \pi$ -pulses (corresponding to $W = 1$) leads to a 99% degree of electron spin synchronization already after a dozen of pulses. However, if the electron spin coherence time is long enough ($T_2 \gg T_R$), an extended train of pulses leads as well to a high degree of spin synchronization, also for $\Theta \ll 1$ ($W \approx \Theta^2/4$).

6.5.3 Tailoring of ensemble spin precession

In this part we test the degree of control over the spin coherence of an ensemble of singly charged dots that can be achieved by periodic laser excitation. For that purpose, a train of two pump pulses is used. Adjustments of the delay between these pulses and their polarization are used to control the shape and phase of the spin coherent signal. The robustness of the mode-locked spin coherence with respect to variations of magnetic field strength is demonstrated.

6.5.3.1 Two pump pulse excitation protocol

Each pump pulse in a train is now split into two pulses with a fixed delay $T_D < T_R$ between them. The results for $T_D = 1.84$ ns are shown in Fig. 6.12(a). Both pumps are circular co-polarized and have the same intensities. When the quantum dots are exposed to only one of the pump pulses (the two upper traces), the Faraday rotation

signals are identical except for a shift by T_D . The signal changes drastically under excitation by the two pulse train (lower trace): Around the arrival of pump 1 the same Faraday rotation response is observed as before in the one-pump experiment. Also around pump 2 qualitatively the same signal is observed with a considerably larger amplitude. This means that the coherent response of the synchronized quantum dot ensemble can be amplified by the second laser pulse.

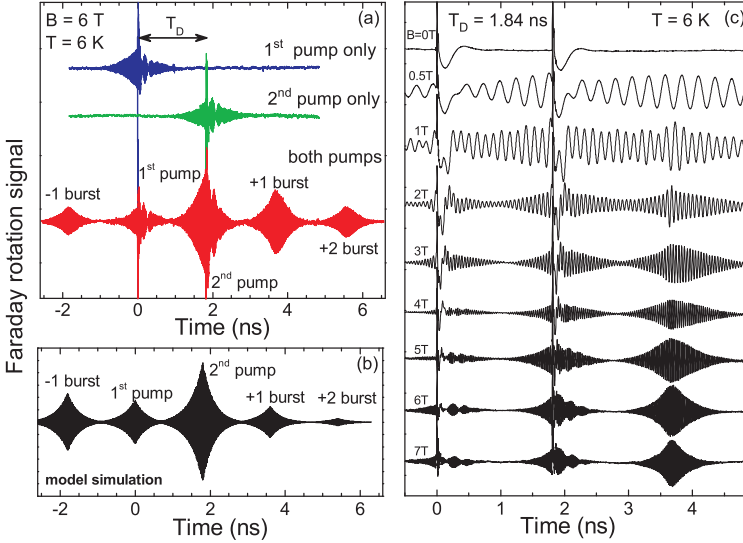


Fig. 6.12 Control of the electron spin synchronization in (In,Ga)As/GaAs quantum dots by two trains of pump pulses with $T_R = 5.2\text{ ns}$, shifted in time by $T_D = 1.84\text{ ns}$. (a) Experimental Faraday rotation signal measured for separate action of the first or the second pump (the two upper curves) and for joint action of both pumps (the bottom curve). The pumps were co-polarized (σ^+). (b) Modeling of the Faraday rotation signal in the two pump pulse experiment with the parameters $\Theta = \pi$ and $\gamma = 3.2\text{ GHz}$. Panel (c) illustrates robustness of the PSC to the variation of magnetic field

Even more remarkable are the echo-like responses showing up before the first and after the second pump pulse. They have a symmetric shape with the same decay and rise times T_2^* . The temporal separation between them is a multiple of T_D . Note that these Faraday rotation bursts show no additional modulation as seen at positive delays when a pump is applied. The reason is that excitons generated in charge neutral dots have already recombined and do not contribute to these signals.

Apparently, the electron spins in the quantum dot sub-ensemble, which is synchronized with the laser repetition rate, have been clocked by introducing a second frequency, which is determined by the laser pulse separation T_D . The clocking results in multiple bursts in the Faraday rotation response. The conditions for this clocking are analyzed in further detail in the next paragraph.

The Faraday rotation bursts due to constructive interference of the electron spin contributions show remarkable stability against variations of the magnetic field in

the range from 1 to 7 T. While the appearance of bursts changes with the field strength (the bursts are squeezed due to the decrease of T_2^* with increasing field), the delay times at which the bursts appear remain unchanged. Also the amplitude of the bursts does not vary strongly in this field range, see Fig. 6.12(c).

6.5.3.2 Signal shaping by changing delay between pump pulses

Figure 6.13 shows Faraday rotation traces excited by a two-pulse train with a repetition period $T_R = 5.2$ ns between the pump doublet. The two pulses have the same intensity and polarization. The delay between these pulses T_D was varied between $\sim T_R/7$ and $\sim T_R/2$. The signal varies strongly, depending on whether the delay time T_D is commensurate with the repetition period T_R , $T_D = T_R/i$ with $i = 2, 3, 4, \dots$, or incommensurate, $T_D \neq T_R/i$. For commensurability the signal shows strong periodic bursts of quantum oscillations only at times equal to multiples of T_D , as seen for $T_D = 1.86$ ns $\approx T_R/7$. Commensurability is also achieved for $T_D = T_R/4 \approx 3.26$ ns and $T_D = T_R/3 \approx 4.26$ ns.

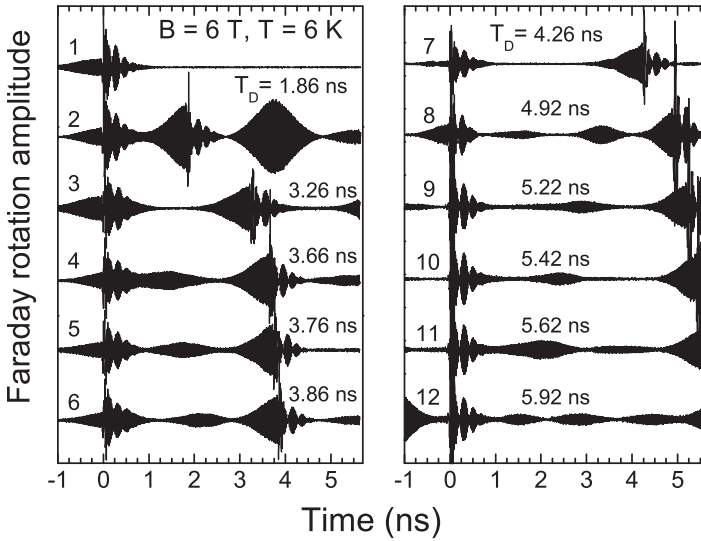


Fig. 6.13 Faraday rotation traces measured as function of delay between probe and first pump pulse at time zero. A second pump pulse was delayed relative to the first one by T_D , as indicated at each trace. The top left trace is measured without the second pump [34]

When T_D and T_R are incommensurate, the Faraday rotation signal shows bursts of spin beats between the two pulses of each pump doublet, in addition to the bursts outside of the doublet. One can see a single burst midway between the pumps for $T_D = 3.76$ and 5.22 ns. Two bursts, each equidistant from the closest pump and also

equidistant from one another, appear at $T_D = 4.92$ and 5.62 ns. Three equidistant bursts occur at $T_D = 5.92$ ns.

Although the time dependencies of the Faraday rotation signals look very different for commensurate and incommensurate T_D and T_R , in both cases they result from constructive interference of synchronized spin precession modes [34]. For a train of pump pulse doublets the phase synchronization condition involves the intervals T_D and $T_R - T_D$ in the laser excitation protocol

$$\omega_e = 2\pi NK/T_D = 2\pi NL/(T_R - T_D), \quad (6.17)$$

where K and L are integers. This condition imposes limitations on the T_D values, for which synchronization is obtained:

$$T_D = [K/(K+L)]T_R, \quad (6.18)$$

which for $T_D < T_R/2$ leads to $K < L$. This phase synchronization condition explains the position of all bursts in the signals from Fig. 6.13. For commensurability, one has $K = 1$ so that $T_D = T_R/(1+L)$. In this case constructive interferences should occur with a period T_D as seen for $T_D = 1.86$ ns ($L = 6$).

For incommensurability the number of bursts between the pulses and the delays at which they appear can be tailored. There should be just one burst, when $K = 2$, because then the constructive interference must have a period $T_D/2$. In experiment, a single burst is indeed seen for $T_D = 3.76$ ns ($L = 5$) and 5.22 ns ($L = 3$), see Fig. 6.13. Two bursts are seen for $T_D = 4.92$ and 5.62 ns, corresponding to $K = 3$ and $L = 5$ and 4 , respectively. Finally, the Faraday rotation signal with $T_D = 5.92$ ns shows three bursts between the pumps, which is described by $K = 4$ and $L = 5$. Thus a good agreement between experiment and theory is established, highlighting the high flexibility of the pump protocol.

6.5.3.3 Polarization control of signal phase

To obtain further insight into the tailoring of electron spin coherence by a two-pulse train, we change from co- to counter-circular polarized pumps. T_D is fixed at $T_R/6 \approx 2.2$ ns. The appearances of the corresponding Faraday rotation signals are similar, as shown in Fig. 6.14(a). Besides the two bursts directly connected to the pump pulses one sees one further +1 burst. The insets show closeups of different bursts. The phase of the spin beats differs by π between the co- and counter-polarized configuration for the pump 1 and +1 bursts. On the other hand, there is no phase difference for the pump 2, -1 and +2 bursts (the last two signals are not shown here). The sign, κ , of the Faraday rotation amplitude in the counter-circular configuration undergoes T_D -periodic changes in time as compared to the constancy in the co-circular case, see Fig. 6.14(b). This demonstrates optical switching of the electron spin precession phase by π in an ensemble of quantum dots.

The observed effect of phase sign reversal is well described by the model [34]. A detailed consideration shows that the modulus of the spin synchronization ex-

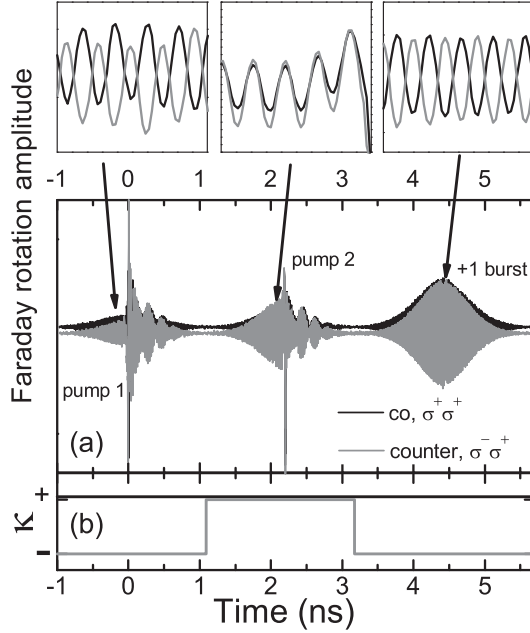


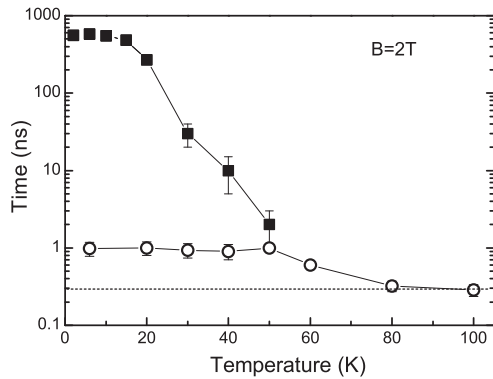
Fig. 6.14 (a) Faraday rotation traces in the co-circular (black line) or counter-circular (gray line) polarized two pump pulse experiments, measured for $T_D = 2.2$ ns, $B = 6$ T and $T = 6$ K. The signal amplitudes look very similar for the two configurations and can hardly be distinguished on the displayed time scale. Therefore the three additional panels give close-ups of the two traces showing the relative sign, κ , of the Faraday rotation amplitudes. κ is plotted in panel (b) vs time [34].

hibits constructive interference with a period $T_R/6$ and changes its sign with a period $2T_R/6$. The relative sign of the Faraday rotation amplitude for the counter- and co-circular cases, $\kappa = \text{sign}\{\cos[\pi t/(T_R/6)]\}$, is in agreement with the experimental data in Fig. 6.14(b).

6.5.4 Temperature dependence of electron spin coherence time

Using the mode-locking technique for measuring the electron spin coherence time, as described in Section 6.5.1, we study the temperature dependence of T_2 . The experimental results are shown by squares in Fig. 6.15 [35]. For the applied experimental conditions ($B = 2$ T) the measured spin coherence time is $0.6 \mu\text{s}$ at $T = 2$ K, and remains about this value for increasing temperature up to 15 K. However, then we find a decrease of T_2 down to $0.25 \mu\text{s}$ at 20 K. At 30 K the coherence time is estimated to be 30 ± 10 ns, and it drops further into the 2 ns range at 50 K, where it becomes comparable to the dephasing time T_2^* . This corresponds to a drop by almost three orders of magnitude over a temperature range of 30 K only. At low temperatures all

Fig. 6.15 Coherence time T_2 (squares) and dephasing time T_2^* (circles) vs. temperature for quantum dot electron spins at $B = 2$ T. Dotted line marks the charged exciton lifetime [35]



spin decoherence mechanisms are frozen except of the hyperfine interaction with the nuclei, which limits the electron spin coherence. However, with increasing temperature the phonons are no longer frozen so that the spin orbit interaction might become relevant. Model calculations, however, reveal that the scattering by phonons is by far too weak to account for the strong drop of coherence time [35]. Instead the drop is tentatively attributed to elastic scattering by fluctuations of the hyperfine field, which can result in a sufficiently strong temperature dependence of T_2 .

The stability against variations of external parameters, like magnetic field and temperature, is a consequence of the mode-locking generation mechanism, which is not fixed to the properties of specific dots, e.g. their spectral energy or spin precession frequency. The periodic pump laser train always selects the proper subsets of dots which satisfy the phase synchronization condition, even for strongly varying experimental conditions.

6.6 Nuclei induced frequency focusing

The three-dimensional confinement of the spins in the QDs protects them from many interactions that in higher-dimensional systems lead to effective spin relaxation [36]. However, strong localization of the electrons increases the electron-nuclear interaction, leading to faster decoherence and dephasing [24, 25]. This problem may be overcome by polarizing the nuclear spins close to 100% [36], but such a high degree of polarization has not been achieved so far.

Therefore, in an ensemble of QDs, fast electron spin dephasing arises not only from variations of the electron g -factor, but also from nuclear field fluctuations, leading to different spin precession frequencies. The dephasing due to the g -factor variations can be partly overcome by the mode-locking effect described in Section 6.5, which synchronizes the precession of specific electron spin modes in the ensemble with the clocking rate of the periodic pulsed excitation laser. Still, there is a significant fraction of dephased electron spins, whose precession frequencies

do not satisfy the phase synchronization condition. This dephased background may represent a severe obstacle if a synchronized dot ensemble shall be used for quantum information applications. However, we have shown that the dephased background can be completely removed by optical pumping on a seconds time scale [37]. The origin is the involvement of the nuclear spin bath interacting with the resident electrons in quantum dots. The nuclei act onto the electron as an effective magnetic field whose component along the external field has to be added to this field when calculating the spin precession frequency of the electron: $\omega = \omega_e + \omega_N$.

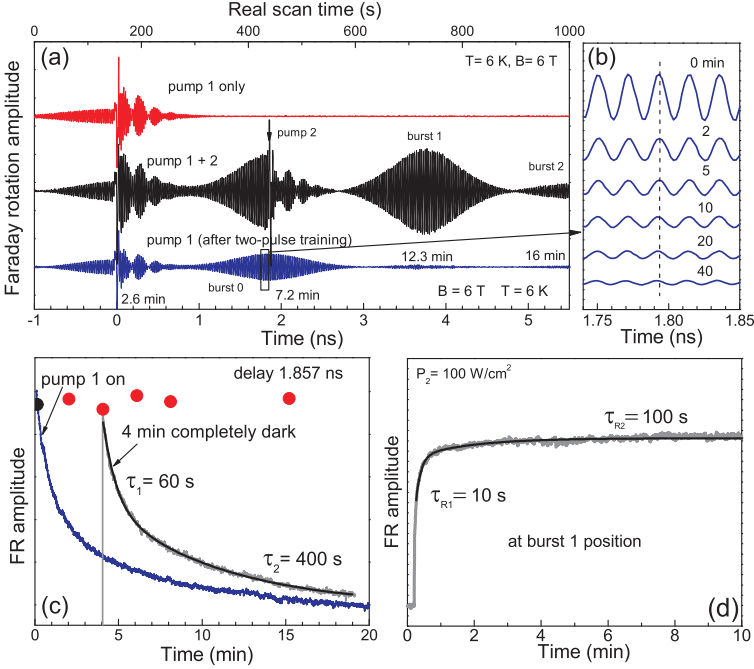


Fig. 6.16 (a) Faraday rotation traces measured on an ensemble of singly-charged (In,Ga)As/GaAs quantum dots. Details of the optical excitation protocol are given in the sketch. The top trace was measured using a train of single pump pulses. The middle trace was excited by a two pump pulse protocol with the second pump delayed by $T_D = 1.86$ ns relative to the first one. The measurement over the whole delay time range took about 20 minutes (top scale). The lowest trace was taken for a single pump pulse excitation protocol with pump 2 closed. Recording started right after measurement of the middle trace. Some times at which the different bursts were measured are indicated. The pump and probe power density were 50 and 10 W/cm². (b) Faraday rotation signals measured over a small delay range at the maximum of 'burst 0' for different times after closing the second pump, while pump 1 and the probe were always on. (c) Relaxation kinetics of the Faraday rotation amplitude at a delay of 1.857 ns after switching off pump 2. Before this recording, the system was treated for 20 minutes by the two pump excitation. The left curve was measured with pump 2 blocked at $t = 0$. The circles show the signal for different times in complete darkness (both pumps and probe were blocked). The second curve shows the decay by the pump 1 and probe illumination after 4 min in dark. $B = 6$ T and $T = 6$ K. (d) rise time at burst 1 position, by switching on the pump 2 [37]

The experiments were done on the same sample containing an ensemble of self-assembled (In,Ga)As/GaAs dots as used in the previous Section. Further, the two pump pulse Faraday rotation technique was applied. Signals measured for single pump and two pump protocols are shown Fig. 6.16(a). The middle trace was recorded after the sample was illuminated for ~ 20 minutes by the pump-doublet train. Immediately after this measurement, the second pump was blocked and a measurement using only the single pump train was started (bottom trace). The times for the delay line to move to the certain position are shown on the bottom curve and top scale. One would expect that blocking of the second pulse in a pump doublet would destroy the periodic burst pattern on a μs time scale according to the electron spin coherence time, T_2 , in these dots (see Section 6.5), so that only the signal around the first pump should remain over the scanned range of pump-probe delays. Contrary to the expectations, the signal shows qualitatively all characteristic for a pump doublet protocol. A strong signal ('burst 0') appears around the delays where the second pump was located. Further signals, denoted 'burst 1' and 'burst 2', also present. Surprisingly, the signal pattern created by the two pulse protocol is memorized over several minutes!

Additional Faraday rotation traces were recorded in a short delay range around 'burst 0' for different times after closing the second pump (Fig. 6.16(b)). A strong signal is seen even after 40 minutes. The decay kinetics was measured at a fixed delay of 1.857 ns (corresponding to the maximum signal) vs the time after switching off pump 2 (left curve in Fig. 6.16(c)). The observed dynamics is well described by a bi-exponential dependence on elapsed time t , $a_1 \exp(-t/\tau_1) + a_2 \exp(-t/\tau_2)$. The decay, however, critically depends on the light illumination conditions. When the system is held in darkness (both pumps and probe are blocked), no relaxation occurs at all on an hour time scale. This is shown by circles in Fig. 6.16(c), which give the initial Faraday rotation amplitude of the burst 0 at the pump 2 position, when switching on only pump 1 and probe after a dark time interval. An example of a decay curve is shown for the dark interval of 4 min. The initial amplitude (presented by the circle) does not change over the dark time. Note, that the rise time of the effect also has a slow component on a minute time scale (Fig. 6.16(d)). This sets the condition for the preparation of the system for about 20 min before any decay measurements.

The observed long memory of the excitation protocol must be imprinted in the dot nuclei, for which long spin relaxation times up to hours or even days have been reported in high magnetic fields [39, 38]. The nuclei in a particular dot must have been aligned along the magnetic field through the hyperfine interaction with the electron during exposure to the pump train. This alignment, in turn, changes the electron spin precession frequency, $\omega = \omega_e + \omega_{N,x}$, where the nuclear contribution, $\omega_{N,x}$, is proportional to the nuclear polarization. The slow rise and decay dynamics of the Faraday rotation signal indicate that the periodic optical pulse train stimulates the nuclei to increase the number of dots for which the electron spin precession frequencies satisfy the phase synchronization condition for a particular excitation protocol. But what is driving the projection of the nuclear spin polarization on the magnetic field to a value that allows an electron spin to satisfy the PSC?

The nuclear polarization is changed by electron-nuclear spin flip-flop processes resulting from the Fermi-contact-type hyperfine interaction [40]. Such processes, however, are suppressed in a strong magnetic field due to the energy mismatch between the electron and nuclear Zeeman splitting by about three orders of magnitude. Flip-flop transitions, which are assisted by phonons, have a low probability due to the phonon-bottleneck [42, 41]. This explains the robustness of the nuclear spin polarization in darkness (Fig. 6.16(c)). Consequently, the resonant optical excitation of the singlet trion becomes the most efficient mechanism in the nuclear spin polarization dynamics. The excitation process rapidly turns "off" the hyperfine field of a resident electron acting on the nuclei, and the field is subsequently turned "on" again by the trion radiative decay. Thereby it allows a flip-flop process during switching without the violation of energy conservation.

The nuclear spin-flip rate for this mechanism is proportional to the rate of optical excitation of the electron, $W(\omega)$. According to the selection rules, the probability of exciting the electron to a trion by σ^+ polarized light is proportional to $1/2 + S_z(\omega)$, where $S_z(\omega)$ is the component of the electron spin polarization along the light propagation direction taken at the moment of pump pulse arrival. Therefore, the excitation rate $W(\omega) \sim [1/2 + S_z(\omega)]/T_R$. For electrons satisfying the PSC, $S_z(\omega) \approx -1/2$, the excitation probability is very low due to Pauli blocking [31]. Due to the very long spin coherence time, T_2 , the excitation rate for these electrons is reduced by two orders of magnitude to $1/T_2$ as compared to $1/T_R + 1/T_2$ for the rest of electrons (in the present experiments $T_2/T_R \approx 200$) [37].

Due to the factor $W(\omega)$, the nuclear relaxation rate has a strong and periodic dependence on ω , with the period determined by the PSC of a particular excitation

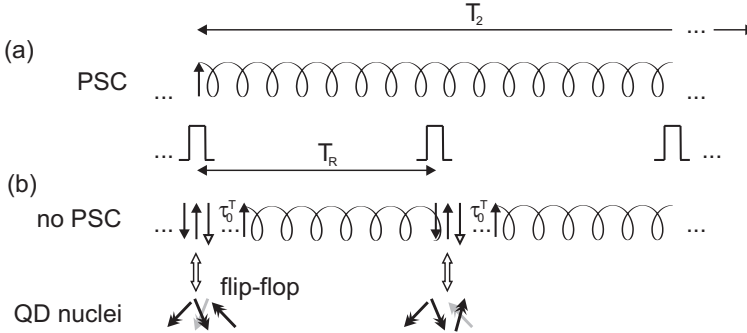


Fig. 6.17 The periodic resonant excitation by a mode-locked circular polarized laser synchronizes the precessions of electron spins whose frequency satisfy the phase synchronization conditions (PSC). At the same time this excitation leads to a fast nuclear relaxation time in quantum dots, which do not satisfy the PSC, via optically stimulated electron-nuclear spin flip-flop processes. The random fluctuation of the nuclear spin modifies the electron spin precession frequency and becomes frozen when this frequency reaches the PSC. (a) Phase synchronized modes do not interact with the pump pulses and therefore electron-nuclear interaction is suppressed within T_2 . (b) For the non-synchronized dots, light is absorbed every 5.2 ns (T_R) and the electron-nuclear flip-flop process becomes possible

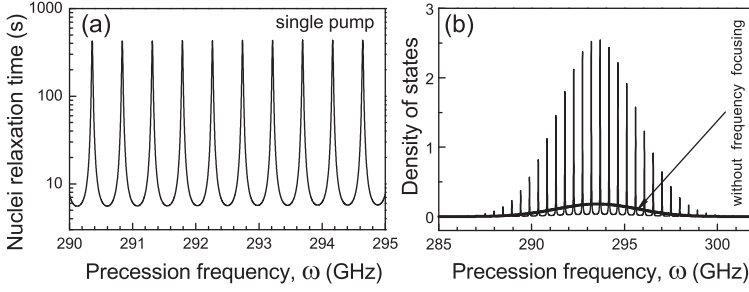


Fig. 6.18 (a) Average spin relaxation time of the As nuclei vs the electron spin precession frequency calculated for the single pump pulse. (b) Density of electron spin precession modes in an ensemble of singly charged dots modified by the nuclei, calculated for the single pump excitation protocols. The thick black line shows the unmodified density of electron spin precession modes due to the dispersion of the electron g -factor in the dot ensemble and due to nuclear polarization fluctuations. Calculations were done for $B = 6$ T, $|g_e| = 0.555$, $\Delta g_e = 0.0037$, $\gamma = 1$ GHz, $T_R = 5.2$ ns, and $T_2 = 3$ μ s [37]

protocol: $2\pi/T_R$ for the single pulse train (Fig. 6.18(a)) and $2\pi/T_D$ for the double pulse train. The huge difference in the nuclear flip rate explains why $\omega_{N,x}$ in each dot tends to reach a value allowing the electron spin to fulfill the PSC. In dots where the PSC is not fulfilled, the nuclear contribution to ω changes randomly due to the light-stimulated nuclear flip-flop processes on a seconds time scale. The typical range $\Delta\omega_{N,x}$ of this contribution to ω is limited by statistical fluctuations of the nuclear spin polarization. For the studied (In,Ga)As dots, $\Delta\omega_{N,x}$ lies on a GHz scale which is comparable with the separation between the phase synchronized modes $2\pi/T_R \sim 0.48$ GHz [37]. As a result, the nuclear contribution occasionally drives an electron to a PSC mode, where its precession frequency is virtually frozen on a minutes time scale. This leads to the frequency focusing in each dot and to accumulation of the dots for which electron spins match the PSC. We mention, that we consider here only the degenerate case for the pump and probe which are resonant with the QD transitions. If one drives the system with off resonant pump pulses, then, dependent on the sign of detuning, the nuclear polarization shifts the precession frequencies to or away of the phase synchronized modes [43]. The frequency focusing modifies the spin precession mode density of the dot ensemble (Fig. 6.18(b)). Without focusing, the density of the electron spin precession modes is Gaussian with a width: $\Delta\omega = [(\Delta\omega_{N,x})^2 + (\mu_B \Delta g_e B / \hbar)^2]^{1/2}$, where Δg_e is the g -factor dispersion. Frequency focusing modifies the original continuum density to a comb-like distribution. Eventually for this distribution the whole ensemble of QD electrons participates in a coherent precession locked on only a few precession frequencies. This suggests that a laser protocol (defined by pulse sequence, width, and rate) can be designed such that it focuses the electron-spin precession frequencies in the dot ensemble to a single mode. This will be shown in Section 6.7.

The focusing of electrons into PSC modes is directly manifested by the Faraday rotation signals in Fig. 6.16(a), as it causes comparable amplitudes before and after the pump pulses. The calculations demonstrate that, without frequency focusing,

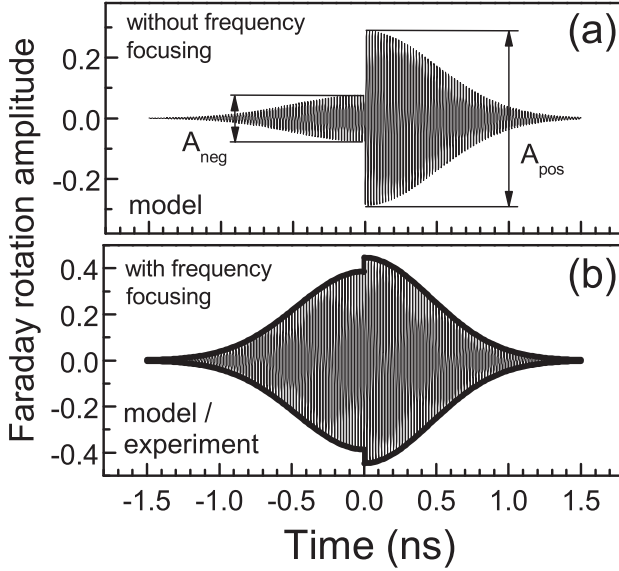


Fig. 6.19 Faraday rotation traces of an ensemble of charged QDs subject to a single pump pulse excitation protocol with pulse area $\Theta = \pi$ at a magnetic field $B = 6$ T. (a) FR traces calculated with the density of electron spin precession modes unchanged by the nuclei. (b) Experimental FR trace with the density of electron spin precession modes modified by the nuclei. It looks almost identical to the theoretical one, which is shown here by thick lines as envelope of fast precessing electron spin beats. Experimental trace is obtained after extracting the contribution of neutral excitons. For the calculation parameters see Fig. 6.18 [37].

the amplitude at negative delays, A_{neg} , does not exceed 30% of the positive delay signal amplitude, A_{pos} (Fig. 6.19(a)). The strong optical pump pulses in the experiment address all quantum dots, and their total contribution should make the Faraday rotation signal much stronger after the pulse than before, when only mode-locked electrons are relevant. However, the nuclear adjustment increases the negative delay signal to more than 90% (Fig. 6.19(b)) of the positive delay signal. The large experimental value of $A_{\text{neg}}/A_{\text{pos}}$ confirms that in our experiment almost all electrons in the optically excited dot ensemble become involved in the coherent spin precession.

Therefore, the nuclei in singly charged QDs exposed to a periodic pulsed laser excitation drive almost all the electrons in the ensemble into a coherent spin precession. The exciting laser acts as a metronome and establishes a robust macroscopic quantum bit in dephasing free subspaces. This may open new promising perspectives on the use of an ensemble of charged QDs during the single electron coherence time T_2 .

6.7 Collective single-mode precession

The nuclei frequency focusing allows us to remove the background of dephased spins by the nuclei. However, the pulsed optical excitation still involves a considerable number of precession modes, as evidenced by the dephasing of spin coherence in the Faraday rotation spectra (Fig. 6.9(b)). In order to reduce dephasing one needs to reduce the number of optically excited synchronized modes.

Let us estimate the number of mode-locked modes that contribute to the FR signal for a particular magnetic field strength. The separation between these modes is $\omega_R = 2\pi/T_R$ (see Fig. 6.11 and Eq. (6.16)) [44]. Consequently, the number of such modes, M , is:

$$M = 2\Delta\omega/\omega_R = \Delta\omega T_R/\pi, \quad (6.19)$$

where $\Delta\omega$ is the half-width dispersion of electron spin precession frequencies in the QD ensemble:

$$\Delta\omega(\Delta g_e, \Delta\omega_{N,x}) = \sqrt{[\mu_B \Delta g_e B / \hbar]^2 + \Delta\omega_{N,x}^2}. \quad (6.20)$$

Here Δg_e is the dispersion of electron g -factors in the ensemble of optically excited dots and $\Delta\omega_{N,x}$ is the nuclear contribution to the dispersion of electron spin precession frequencies for each specific dot. The magnitude of $\Delta\omega_{N,x}$ is determined by statistical fluctuations of the nuclear spin polarization projection onto the magnetic field in the dot volume [24]. For our dots $\Delta\omega_{N,x} = 0.37$ GHz. This value was obtained from the amplitude of the random nuclei fluctuation field of 7.5 mT measured by a technique described in Ref. [45].

Equations (6.19, 6.20) define a clear strategy for achieving the single mode precession regime in a QD ensemble. The number of mode-locked modes can be reduced: (a) by minimizing Δg_e , (b) by reducing T_R , (c) by decreasing B , and (d) by decreasing $\Delta\omega_{N,x}$.

(a) Generally, the dispersion Δg_e in a QD ensemble is connected to variations of dot shape and size. For (In,Ga)As dots a systematic dependence of the electron g factor on energy of the band edge optical transitions has been observed [6]. Δg_e can then be controlled by the laser spectral width, which is inversely proportional to the pulse duration. However, as one is interested in fast spin initialization, the duration should not exceed ~ 10 ps (spectral width ~ 0.1 meV). Otherwise the efficiency of spin polarization initialization drops when the pulse duration becomes comparable with the times of electron or hole spin precession and electron-hole recombination.

(b) A reduction of the repetition period is generally limited by the trion decay time and the time scale that multiple coherent operations would require. For practical reasons, to observe the single mode regime, T_R should be longer than the ensemble dephasing time of a few ns [6].

(c) A reduction of the magnetic field strength is possible to an extent that it is still considerably larger than the randomly oriented effective field of the nuclei. Otherwise the nuclei would induce fast dephasing [25, 24]. In our dots the random nuclear fluctuation field has an amplitude of about 7.5 mT [45]. Also, to conserve

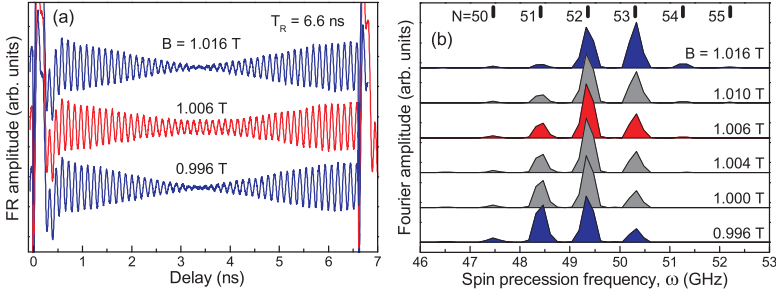


Fig. 6.20 (a) Faraday rotation spectra recorded around $B = 1$ T. (b) Fourier transforms extracted from FR signals. Positions of phase-synchronized modes are marked by vertical lines [44]

nuclear spin polarization, which is needed for frequency focusing, the magnetic field should exceed the hyperfine field of the electron acting on the nuclei (Knight field), which is about 1–3 mT in our dots.

Let us pick up mostly on the method (c). The experiments shown so far were done at field strengths of a few Tesla. In a first step we therefore reduced the field strength to about 1 T. Figure 6.20(a) shows the corresponding Faraday rotation traces. The pump pulses hit the sample at times $t = 0$ and 6.6 ns, so that also tool (b), reducing T_R from 5.2 to 6.6 ns, was applied. Over a narrow range of magnetic fields the Faraday rotation traces undergo strong modifications although the oscillation frequencies appearing in them do not change significantly. At $B = 0.996$ and 1.016 T the signal amplitude shows a strong decay after the first pulse, hits a node in the middle between the pump pulses and afterwards increases symmetrically towards the second pump pulse. However, at the intermediate field of 1.006 T, deviating by 10 mT only from the two other traces, the signal decay after the first pulse is weaker and, in particular, it does not show a node. This non-monotonic behavior of the Faraday rotation signal with B is repeated every 0.02 T. This is also consistent with the distance between synchronized modes in B -field equivalent: $\Delta B = \hbar\omega_R / (|g_e|\mu_B) = 20$ mT for $T_R = 6.6$ ns and $|g_e| = 0.54$.

The precession frequency spectra can be derived from the Faraday rotation traces by Fourier transforms. The Fourier spectra in Fig. 6.20(b) confirm that the Faraday rotation signal around 1 T is contributed by 2–3 precessional modes, neglecting further, rather weak side modes. With increasing magnetic field the center of the contributing precession frequencies shifts to higher values with respect to the discrete mode-locked frequency spectrum, $\omega = 2\pi N/T_R$, where $N = 50$ –55 around 1 T. This shift changes the relative contribution of the different modes to the Fourier spectra. If for a magnetic field two strong modes of the same weight dominate the spectrum, the Faraday rotation signal shows a node between the pump pulses, see $B = 0.996$ and 1.016 T. For the intermediate field of $B = 1.006$ T, the Faraday rotation signal is determined by a central mode which is accompanied, however, by strong symmetric satellites. Therefore, the FR signal has considerable amplitude between the pump pulses.

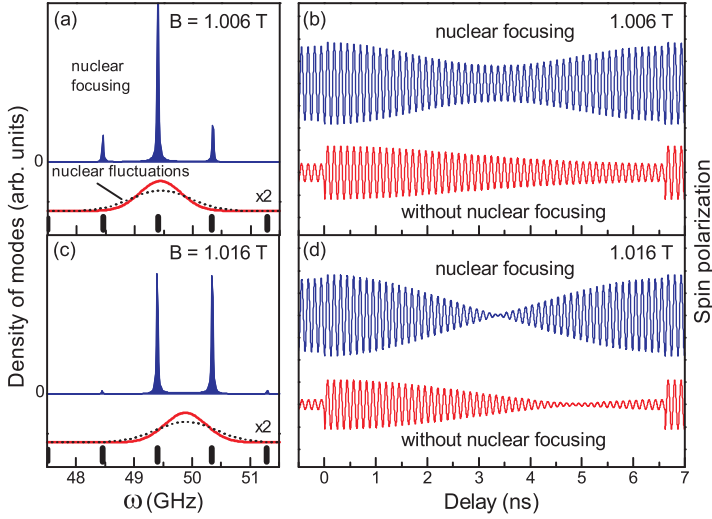


Fig. 6.21 Calculations of the density of electron spin precession modes and corresponding FR signals at $B = 1.006$ and 1.016 T. Panels (a),(c) show the density of modes created by g -factor dispersion (solid line), by nuclear fluctuations (dot line) and by nuclei induced frequency focusing. Positions of phase-synchronized modes are marked by vertical lines. Panels (b),(d) show the spin polarization which is proportional to the FR signal magnitude, calculated with and without frequency focusing. Parameters for calculations: $|g_e| = 0.556$, $\Delta g_e = 0.004$, $\Delta \omega_{N,x} = 0.37$ GHz, and pump pulses with π -area [44]

In Fig. 6.21 we show the theoretical simulations for the mode density and the corresponding FR spectra created by a train of π pulses with repetition period $T_R = 6.6$ ns at $B = 1.006$ and 1.016 T. The calculations [44] are in good agreement with the experimental data. For comparison one can also see the corresponding dependencies without nuclei induced frequency focusing. The significant deviation from the experimental observations underlines the importance of the nuclear contribution in the mode-locking.

Figure 6.22(a) presents the magnetic field dependence of precession frequency dispersion $\Delta\omega(\Delta g_e, \Delta\omega_{N,x})$ and mode number M calculated for our dots under the applied experimental conditions. For $B < 0.8$ T, we estimate $0.75 < M \leq 1$, giving a lower limit for the number of mode-locked frequencies in the FR signal. In this range the Δg_e contribution to $\Delta\omega$ (dash-dotted line) is smaller than the nuclei induced dispersion (dashed line). The total dispersion is therefore approximately equal to $\Delta\omega_{N,x}$, which is independent of magnetic field. This results in periodic switching between almost pure single and double mode regimes for $B < 0.8$ T. Whether one or two modes fall within the dispersion can be adjusted by the magnetic field, which shifts the spectrum of optically excited electron spin precession frequencies in the QD ensemble relative to the spectrum of phase synchronized modes.

The magnetic field dependence of the FR amplitude midway between the pump pulses (at a delay of 3.3 ns, where the node is observed) oscillates symmetrically

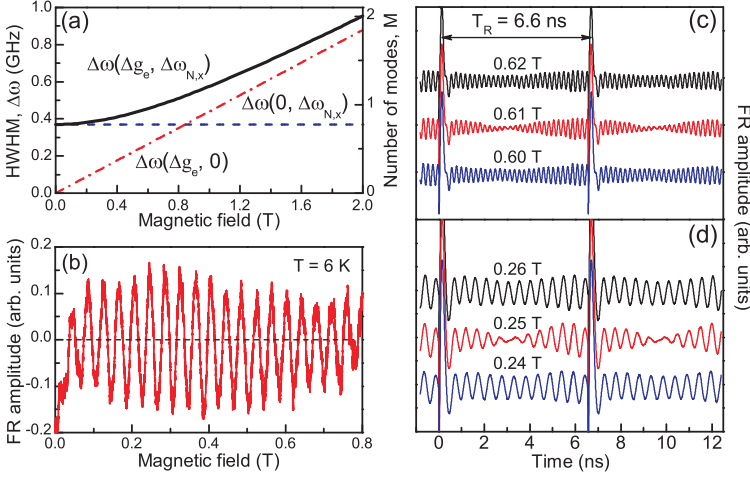


Fig. 6.22 (a) Magnetic field dependence of dispersion of electron spin precession frequencies, $\Delta\omega(\Delta g_e, \Delta\omega_{N,x})$, (left scale) and of number of mode-locked frequencies (right scale) contributing to the FR signal (solid line). Dashed line shows the nuclear contribution to the dispersion, $\Delta\omega_{N,x}$, in our QDs. The contribution by the electron spin g -factor dispersion is shown by dash-dotted line. Parameters are the same as in Fig. 6.21. (b) Magnetic field dependence of Faraday rotation signal at 3.3 ns delay for pump and probe powers of 5 W/cm² each. (c), (d) Faraday rotation signals recorded at lower magnetic fields.

around zero value illustrating the mode switching (Fig. 6.22(b)). It is partly contributed by the change of central precession frequency by the field scan, as the harmonic oscillation of spin precession is tuned through the fixed delay. But the dominating effect comes from the overall amplitude oscillation due to mode interference. The strongest signal in panel (b), either minimum or maximum, is reached for an odd number of modes, while zero signal is caused for an even mode number. Magnetic fields larger than 0.8 T increase the dispersion of spin precession frequencies, see Fig. 6.22(a), and allow $\Delta\omega$ to cover more than three mode-locked frequencies. This increases the amplitude of side modes significantly, as seen in Fig. 6.20(b) for $B = 1$ T, and, consequently, leads to considerable dephasing.

To address experimentally the single mode regime we apply weaker magnetic fields in order to minimize the contribution of Δg_e . The experimental results shown in Fig. 6.22(c) and (d) were measured around 0.61 and 0.25 T, respectively. This clearly shows a node in the middle (for 0.61 and 0.25 T) and virtually no decay between the pump pulses for the fields with +10 or -10 mT relative to the "node" field. Our model calculations show that under this condition about 95% of precession frequencies are focused into the single mode [44].

(d) For a true single mode regime the condition: $\Delta\omega_{N,x} < 2\pi/T_R$ should be satisfied. This can be reached by increasing the dot size, because $\Delta\omega_{N,x}$ is controlled by the statistical fluctuations of the nuclear spin polarization in the dot volume, V , given by $\Delta\omega_{N,x} \sim 1/\sqrt{V}$. Otherwise, quantum dots with other nuclear composition, e.g. with nuclei having smaller nuclear spins, might be studied. The results

presented in last three Sections show that the shortcomings which are typically attributed to quantum dot spin ensembles may be overcome by carefully designed laser excitation protocols. The related advantages are due to the robustness of the phase synchronization of the quantum dot ensemble: (i) a strong detection signal with relatively small noise; (ii) changes of external parameters like repetition rate and magnetic field strength can be accommodated for the phase synchronization condition due to the broad distribution of electron spin precession frequencies in the ensemble and the large number of involved quantum dots. Additionally, this allows us to drive an entire ensemble of electron spins (about a million spins), confined in singly charged quantum dots, into coherent single-mode precession.

6.8 Ultrafast optical spin rotation

One of the DiVincenzo criteria for the realization of the quantum computing is the ability to generate the logic gate [1], which in our case is presented by the spin with the possibility of arbitrary spin rotation. Another essential point is how fast one can realize these rotations. This operational speed should be much faster than the spin coherence time, T_2 . The quantum information theory set the lower limit for this requirement: it should be at least on the order of 10^6 operations within the coherence time [46, 47]. As we already know from Section 6.5, the spin coherence time in the studied (In,Ga)As quantum dots is about $3 \mu\text{s}$. Thus, for optical control on the picosecond timescale, about 10^6 operations could be carried out during that coherence time. Spin rotations have been obtained for gated GaAs QDs using RF-fields, but such manipulations are slow (on the order of nanoseconds), with times comparable with the spin coherence time [48]. In an all-optical experiment on shallow interface fluctuation QDs, weak confinement precluded full unitary spin rotations [49]. Evidence for spin rotations was reported for a single GaAs interface fluctuation quantum dot using very intense optical excitation needed for spins to be rotated by precession about the effective laser magnetic field [50]. More convincing rotations were reported for a single InAs self-organized QD [51]. In general, optical coupling to a spin in a single quantum dot is weak, and thus spin rotations on single QDs give weak responses. This generally requires many optical pulses to obtain sufficient fidelity, and it can give additional dephasing.

An ensemble of QDs has the advantage of increasing the optical coupling and can decrease the loss of information from the loss of a few spins. However, ensemble approaches typically are hampered by the inhomogeneities in QD properties, particularly in the spin splitting g -factor, and lead to dephasing of the spin precession about a magnetic field [25, 24]. In Section 6.6 we show that the electron spin coherence in ensemble of QDs can be focused to a single mode regime. We use this regime here for optical manipulation of the electron spins.

For further consideration of the coherent manipulations of the spin it is convenient to use the Bloch sphere representation. Figure 6.23(a) presents the time evolution of the single spin in a magnetic field after the optical orientation with the

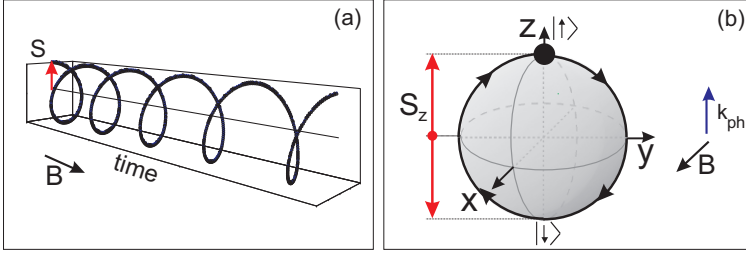


Fig. 6.23 (a) Larmor precession of a spin S about the magnetic field, B . (b) Precession of the spin around the magnetic field is represented on a Bloch sphere by the black trajectory with arrows. The measured ellipticity is proportional to the projection of electron spin polarization on the z -axis (S_z). k_{ph} is the wave vector of the pump light generating electron polarization along z -axis [52]

laser pulse. The optical pulse of circular polarization excites the spin along z -axis, and the magnetic field is directed along the x -axis. The same spin precession can be presented as the rotation of the spin vector on the Bloch sphere in the $z - y$ plane, shown as black line with arrows in Fig. 6.23(b). Here the magnetic field is directed along the x -axis and the circular polarized light orients the spins along the z -axis, so that the North pole is defined as the spin up state, $|\uparrow\rangle$, and the South pole as the spin down state, $|\downarrow\rangle$. Then the y -axis in positive direction is the superposition of both: $(|\uparrow\rangle + |\downarrow\rangle)/\sqrt{2}$.

Experimentally precession of spins is measured by ellipticity of a probe laser and is proportional to the spin polarization along the optical z -axis [52]. We used two synchronized Ti:Sapphire lasers each emitting pulses of 1.5-ps duration with a separation of $T_R = 5.2$ ns. The pump and probe beams create and test the spin polarization and are originated from the same laser. The second laser delivers the pulse for the optical control of the spin polarization created by the pump pulse. The ellipticity with no control laser is shown by the bottom (thick black) trace in Fig. 6.24(a) vs pump-probe delay. The precession of the spins exhibits a weak dephasing arising from the distribution of precession frequencies around the main mode $2\pi N/T_R$ with $N = 30$ at $B = 0.29$ T, see Section 6.7.

The control laser pulses in the experiments shown in Fig. 6.24 have the same photon energy as the pump pulses. Therefore, unlike the case of RF-control [48], the optical control pulse can not only rotate the electron spin coherence generated by the pump, but by itself generate the electron spin coherence. This generation is not desirable for control pulses and in order to avoid it we use the control with the 2π pulse area. Such pulses move the (optical) polarization through a full rotation (a Rabi flop, see Section 6.4) to the optically excited state and back to the ground state. Then, a phase φ is induced to the ground state. Due to selection rules, this is a relative phase between the spin states pointing along $+z$ and $-z$, and it is equivalent to a rotation about the z -axis by an angle φ (for more details see Chapter 5). To understand the actions of the 2π control pulse lets again consider the Eq. (6.10), which derives from the pulse action on the superposition of $\alpha |\uparrow\rangle + \beta |\downarrow\rangle$. For the $\Theta = 2\pi$ pulse this takes the form of: $\alpha |\uparrow\rangle - \beta |\downarrow\rangle$, what is equivalent to the rotation

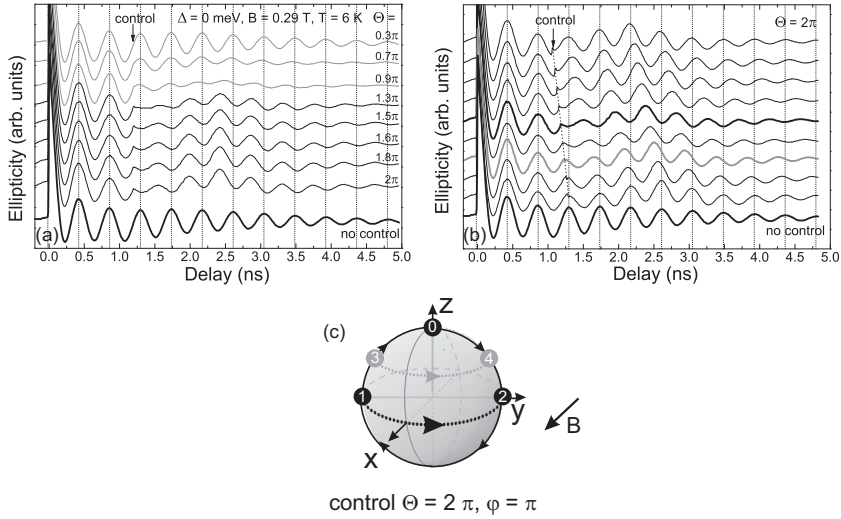


Fig. 6.24 (a) Measurements of ellipticity of the total spin of an ensemble vs. delay time between pump and probe. The pump and probe energies are degenerate with control. The bottom (thick black) trace is without a control laser. The control area is varied to identify the 2π pulse via a phase shift of π of the spin relative to no control. (b) $\Theta = 2\pi$ vs pump-probe delay with the control at varying delays after the pump. The bottom trace is again without a control laser. (c) The effects of the control delay sketched on the Bloch sphere. The two trajectories have grey scaling corresponding to the thick grey and black traces of panel (b) [52]

on the angle $\varphi = \pi$ around the z -axis on the Bloch sphere (see Fig. 6.24(c)). The spin is oriented along the z -axis by the pump, as shown by the black dot marked with zero on the sphere (Fig. 6.24(c)). It precesses about the magnetic field until it is hit by the control, which should induce a $\varphi = \pi$ rotation about z -axis. Then it continues its precession.

The control pulse hits the sample 1.2 ns after the pump, when the spin vector is along the $-y$ axis, so that the π rotation should bring it to $+y$. Pulses with area $\Theta < \pi$ move some of the state to the excited trion level without returning it to the electron spin subspace. Then the spin amplitude is reduced while its phase remains unchanged (top three curves in Fig. 6.24(a)). For $\Theta > \pi$, part of the state is returned to the spin subspace, so that the phase of the spin changes but the amplitude is reduced compared to the case without control (next four curves in Fig. 6.24(a)). The last trace with control in Fig. 6.24(a) is for $\Theta = 2\pi$ control pulse, and it has the maximum amplitude among the cases with control pulses. It shows also clearly the phase change by π in the signal before and after the control pulse arrival. Its amplitude is somewhat reduced from the no control case due to inhomogeneities of dipole moments.

Next we fix $\Theta = 2\pi$ and vary the delay τ between pump and control. This allows the spin to precess in the $y-z$ plane for a time τ (Fig. 6.24(b)). The first and the last traces with control in Fig. 6.24(b) correspond to applying the control pulse when the

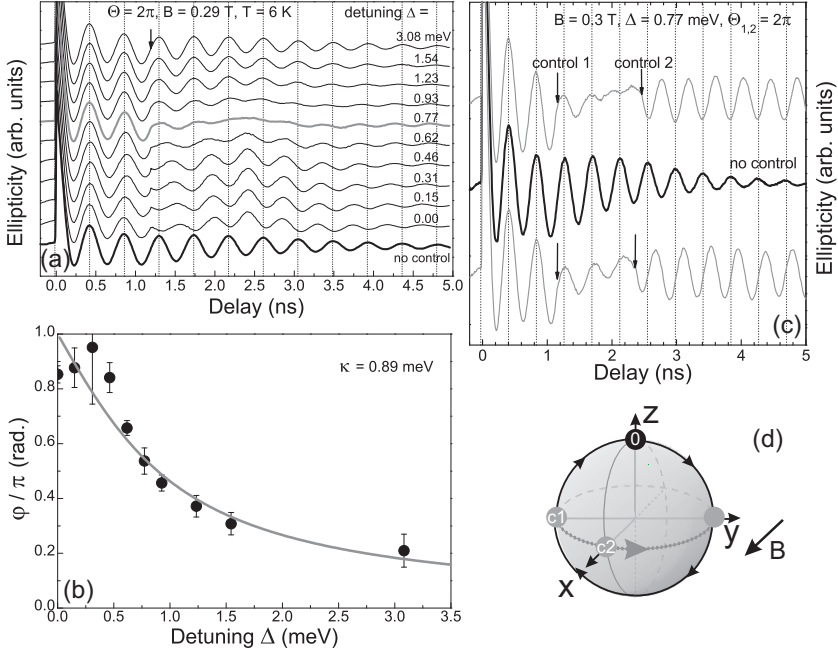


Fig. 6.25 (a) Measurements of ellipticity for varying detunings between control and pump energies. Control pulse intensities are $\Theta = 2\pi$, and the delays between the pump and control pulses were chosen so that the spin is along the $-y$ direction when the control is applied. (b) Symbols give the dependence of rotation angle on the detuning between the control and pump from the data in panel (a). Grey line gives the theoretical result $\varphi = 2 \arctan(\kappa / \Delta)$ from Refs. [54, 53] (see also Chapter 5). (c) Ellipticity measurements with two control pulses with varying temporal separations between them. The two control pulses have area of 2π and the same energy. Their detuning from the pump photon energy is 0.77 meV so that each of them would give a $\pi/2$ rotation about the z -axis, as in the single control case. Together they have the effect of a single π rotation about the z -axis. (d) Bloch sphere depiction of the two-control experiment. The points marked c1, c2 are where the first and second control pulses hit the spin, respectively [52]

spin points in the z direction, in that case it has basically no effect on the spin. For all other spin orientations the control pulse gives a π rotation about the z -axis, after which the spins continue to precess. Figure 6.24(c) shows control induced rotations for two orientations. For the thick black trace the control pulse was applied when the spin points in the $-y$ direction. After the control, it is completely out of phase with the reference trace.

The full control of the coherent spin state suggest the possibility to address the spin to any point of the Bloch sphere. This can be realized by rotations at arbitrary angles about two orthogonal axis. In our case rotation about x -axis is provided by the Larmor spin precession about the external magnetic field $B \parallel x$. Rotation about z -axis can be provided by the 2π control pulse in order to prevent generation of additional spin polarization. Spin rotations of varying angles can be obtained by changing the energy detuning of the control pulse from the optical resonance while

keeping its area at 2π . Figure 6.25(a) gives a series of traces with the detunings (Δ) between 0 to 3.08 meV. We choose the arrival time of the control so that the net spin polarization is along $-y$, in the equatorial plane of the Bloch sphere. Then the effect of rotation by the control becomes most prominent in the phase and amplitude of the ellipticity signal. The angle of spin rotation φ decreases from π to nearly zero as the detuning is increased from zero to 3.08 meV (Fig. 6.25(a)).

For angles of rotation $\pi/2 < \varphi < \pi$, the spin polarization immediately after the control has a component along $+y$ and a nonzero component along the magnetic field. Then the phase of the signal is opposite to that of the reference (Fig. 6.25(a) for detunings < 0.77 meV). For $\varphi < \pi/2$, the y component is still negative right after the control, and the oscillations are in phase with the reference signal. The angles of rotation are obtained from the ratio of the (signed) amplitude over the amplitude of the reference at the maximum signal at 2.4 ns. It is shown as a function of detuning in Fig. 6.25(b). This angle is in excellent agreement with the theoretical expression, which is derived analytically for pulses of hyperbolic secant temporal profile $\text{sech}(\kappa t)$ and gives $\varphi = 2\arctan(\kappa/\Delta)$, where κ is the bandwidth of the pulse [54, 53] (see also Chapter 5).

More than one control pulses can be implemented in the spin manipulation protocol. It is illustrated by Fig. 6.25(c) where two 2π control pulses hitting the sample at different delays are used. Each of this pulse provides the spin rotations by angle $\pi/2$, as the control energy is detuned from the pump one by $\Delta = 0.77$ meV. The first control is applied when the spin is in the $-y$ direction, rotating the spin into the $+x$ direction, along the magnetic field, where it can stay without precession. The second control rotates the spin back into the $y-z$ plane in the $+y$ direction. The efficiency of this action does not depend on the time delay between the two control pulses (surely it should not exceed the spin dephasing time) as one can see from the upper and lower signals in Fig. 6.25(c). For the lower signal the second control is applied when the reference signal (no control) is pointing in the $-z$ direction, which results in a phase difference of $\pi/2$ between the two controls and the reference. In the upper case the second control is applied when the reference spin is in the $-y$ direction, resulting in a phase difference of π between the two-control case and the reference.

Realization of the ultrafast optical spin rotation by means of short laser pulses allows us to perform an all-optical spin echo experiment on ensemble of electrons confined in quantum dots. The spin echo is well known techniques in NMR experiments [55, 56], which is widely used to study spin dynamics in inhomogeneous spin ensembles. Until very recently microwave radiation has been used for spin rotation in the spin-echo technique, which restricts its ultra fast realization. Schematically the all-optical spin echo experiment is shown in Fig. 6.26(a). Spin coherence of the electron ensemble is generated by a π pump pulse. The spins precess in external magnetic field with different frequencies, which causes partial dephasing of the spin coherence. After the time interval τ a 2π control pulse is applied. It has the same photon energy as the pump pulse and therefore rotates spin about z -axis by an angle π . As a result for further precessing the slower spins are moved ahead of the faster spins and all spins refocus at 2τ time moment giving rise to the maximum

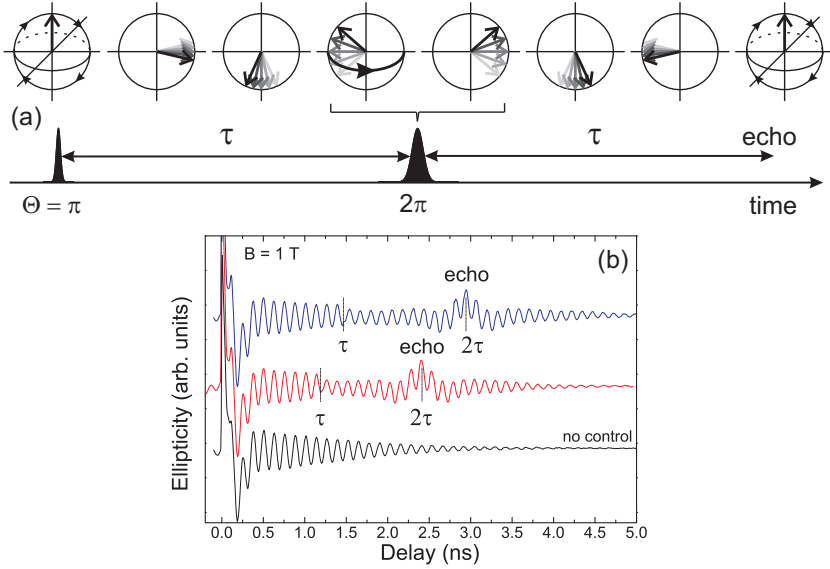


Fig. 6.26 (a) Sketch of spin echo formation vs. time in an optically controlled inhomogeneous spin ensemble. The spins are initialized along z -axis by a pulse of area π after which they precess about the external magnetic field with different frequencies. At time τ a control pulse of area $\Theta = 2\pi$ and zero detuning rotates the spins about the z -axis by an angle π . After 2τ the spins come into phase again and an echo builds up. (b) Experimental demonstration of a spin echo for two time delays of 1.47 and 1.20 ns between pump and control pulses [52]

of the Faraday rotation signal shown in Fig. 6.26(b). In the regime of spin-echo experiments we are able to increase the dephasing time of the ensemble, T_2^* , as seen by comparing the amplitude after 2τ of the rotated spins to the reference. In principle, these π rotations also can extend the T_2 time of the individual spins by decoupling them [57] or disentangling them [58, 59] from the nuclei. To conclude, we have demonstrated a robust, optically controlled single spin rotation gates on the picosecond timescale in quantum dot ensembles. These gates could provide the basis for single-qubit operations, and thus are an important step forward in spin-based quantum dot implementations for quantum information. The present quantum dot experiments were done on ensembles, and they give information about the control of spins at the single dot level. The fidelity of the coherent operations could be affected by dephasing in the quantum dots owing, for example, to decay of optically inactive states or many-body effects, as well as to the effects of inhomogeneities in dipoles or transition energies.

6.9 Conclusions

Dephasing of spin coherence was one of the serious arguments against the functionality of devices containing ensembles of QDs, even for the case when spin coherence in individual dots is held for a long time. This limitation is absent in single dot devices, but there are many factors which make this system difficult, even for basic studies. Among them: The difficulty in controllable growth of self-organized dots with specified energy and with low dot density; very low signal intensity; systematic noise problems in single-electron devices. With mode-locking, we are able to exploit the long spin coherence time of single QDs, but use all advantages of working with a dot ensemble: strong signal; stability of device is not related anymore to the stability of single dot; noise parameters are much better; high temperature stability. The discussed mechanisms raise the question if the properties of QD ensembles make them optimal for quantum coherent devices. A sizable distribution of the electron g -factor is good for mode-locking, as the PSC is fulfilled by many QD subsets, leading to a strong spectroscopic response. Further, it gives some flexibility when changing, for example, the laser protocol (e.g. wavelength, pulse duration or repetition rate) by which the QDs are addressed, and therefore changing the PSC, as the ensemble can offer other QD subsets for which the single dot coherency can be recovered. Additionally, we have demonstrated that mode-locking, combined with nuclei-induced frequency focusing, allows us to drive an entire ensemble of electron spins, into coherent single-mode precession. Therefore, this regime will be very useful for studying various coherent phenomena, such as electromagnetically induced transparency or control-NOT gate operations.

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Part III
**Novel systems for coherent spin
manipulation**

Chapter 7

Optically controlled spin dynamics in a magnetically doped quantum dot

Doris E. Reiter, Tilmann Kuhn and Vollrath M. Axt

Abstract The optically induced spin dynamics in a single quantum dot doped with a single Mn atom are studied theoretically both for the case of a magnetic field applied in Faraday configuration and in Voigt configuration. When the magnetic field is applied in Faraday configuration, the z -component of the angular momentum remains a good quantum number. We show that using a series of ultra short laser pulses manipulating both heavy and light hole excitons allows us to coherently switch the Mn spin from a given initial state into all other spin eigenstates on a picosecond time scale. By modifying the pulse sequence coherent superposition states can be prepared as well. Possible detection schemes are discussed. When the magnetic field is applied in Voigt configuration, the ultrafast optical excitation of an exciton changes the direction of the effective magnetic field acting on the Mn spin on a femtosecond time scale. This induces a precession of the Mn spin which can be efficiently controlled by the application of additional optical pulses.

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7.1 Introduction

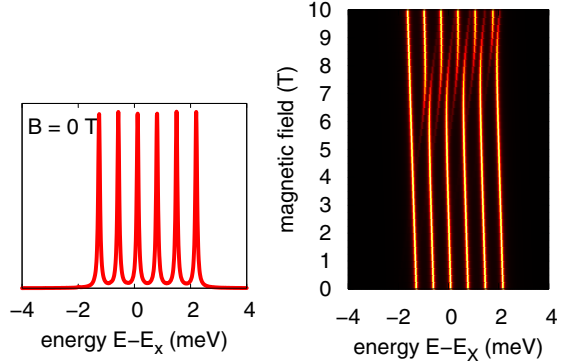
The coherent manipulation of single spins opens the doorway for a variety of applications in quantum information processing and quantum computing [1]. In semiconductor-based systems quantum dots (QDs) are favorite candidates for spin control because of their discrete energy level structure which reduces dephasing and relaxation mechanisms and allows one to selectively address certain quantum states [2, 3, 4]. Many fascinating spin control experiments on the electron spin [5, 6] or the hole spin [7] have shown a good controllability, but also the influence of various dephasing mechanisms has been seen. Impurities embedded in a semiconductor matrix often couple even less to their surroundings and are therefore good candidates for very long spin coherence times. Semiconductors doped with magnetic impurities referred to as *diluted magnetic semiconductors* or *semimagnetic semiconductors* are nowadays widely used in the field of semiconductor spin physics [8], because they combine the advantages of semiconductors with magnetic materials and thus open up a variety of new perspectives in the growing field of spintronics. Indeed, diluted magnetic semiconductor quantum dots have been proposed for applications such as an exciton-controlled magnetization [9, 10] or an optical switching device [11]. A particularly interesting regime is reached when the QD is doped with a single magnetic ion because in this case we have two interacting quantum systems, the quantum dot exciton and the spin of the magnetic ion, both with a discrete spectrum. Such a system has been fabricated and extensively investigated in the past few years by the group in Grenoble, who succeeded in doping a single CdTe QD with a single Mn atom [12]. In the photoluminescence (PL) spectrum of such a dot the exciton line splits into a set of six equidistant lines with essentially equal intensities, showing clearly the influence of the exchange interaction between the exciton and the Mn atom [12, 13]. More recently also an InAs QD has been successfully doped with a single Mn atom [14]. Here a more complicated PL spectrum consisting of five lines with different intensities has been observed. Both types of photoluminescence spectra have been conclusively interpreted by models using a Heisenberg-type exchange interaction between the magnetic ion and the electrons and holes in the QD [15, 16]. The basic difference seen in the PL spectra of II-VI and III-V materials is due to the fact that in the former Mn is an isoelectronic impurity while in the latter it acts as an acceptor. In this chapter we will restrict ourselves to the case of a Mn-doped CdTe quantum dot. Extensive studies of the PL spectra of a Mn-doped CdTe QD under magnetic fields both in Faraday and Voigt configuration revealed many interesting aspects related, e.g., to phenomena like valence band mixing and electron-hole exchange interaction in the dots [17, 18, 19]. In particular they also showed a large variability of the properties depending on microscopic details of the structure like the QD geometry, the presence of strain fields and the exact position of the Mn atom in the QD. By an external bias the charge state of the dot has been controlled in a Schottky structure [20, 21], changing the PL spectra to a set of 11 lines. Such structures have been proposed for single electron transistor applications [22]. For any application based on the different orientations of the Mn spin in the QD it is essential to be able to selectively prepare this spin in a well-defined state, e.g., an

eigenstate of its z -component, and to switch in a controlled way between such states. A scheme for such a control based on selective optical pumping has been proposed [23] and recently a controlled preparation of Mn spin states has been demonstrated experimentally in single [24] and in double QDs [25]. In all these studies the spin transfer was based on incoherent relaxation or transfer processes resulting in typical time scales of the order of several nanoseconds. This time scale is sufficient to prepare the system in a Mn spin eigenstate since recent experiments have shown life times of the Mn spin of the order of tens of nanoseconds [26]. The Mn spin with its six orientations, however, is also an interesting candidate for applications in the field of quantum information processing. Recently it has been shown that quantum systems with a higher dimensional Hilbert space than the usual two-level systems (qubits) might be advantageous for the realization of quantum gates [27]. For quantum computing applications coherent superposition states are of central importance. Such states are usually not generated by incoherent processes. In this chapter we will present a scheme based on the coherent light-induced dynamics of the exciton-Mn system resulting in switching times in the picosecond regime [28]. We will show that by this scheme both Mn spin eigenstates as well as superposition states can be well prepared and a switching between these states is possible by using sequences of ultrafast laser pulses generating and manipulating the QD excitons. The paper is organized as follows: After a discussion of the quantum dot model (Sec. 7.2), the elementary step of such a coherent switch of the Mn spin is discussed (Sec. 7.3), which is then extended to different switching protocols (Sec. 7.4). Finally the control of the Mn spin in a transverse magnetic field is discussed (Sec. 7.5).

7.2 Model System of a single dot doped with a single Mn atom

To describe a single CdTe quantum dot doped with a single Mn atom we follow the model of Fernández-Rossier [15], which successfully explained the observed PL spectra [12]. In the model the electronic structure of the quantum dot is reduced to the lowest conduction band state which can be occupied by two electrons with spin $S^e = \frac{1}{2}$, $S_z^e = \pm \frac{1}{2}$ and the uppermost valence band state, which is typically a heavy hole (HH) state and thus can be occupied by two holes with total angular momentum $S^h = \frac{3}{2}$, $S_z^h = \pm \frac{3}{2}$. For reasons which will become clear below we extend this model by including also the highest state in the light hole (LH) band which can accommodate two holes with angular momentum $S^h = \frac{3}{2}$, $S_z^h = \pm \frac{1}{2}$. The LH states are assumed to be 40 meV below the HH states. The strength of valence band mixing due to asymmetry or strain [29] strongly depends on the individual quantum dot and is neglected in our case. From these states we can form 15 pair states $|X\rangle$ consisting of zero, one, or two electron hole pairs, namely the ground state $|0\rangle$, where no exciton is present, four HH single exciton states $|H \pm 2\rangle$ and $|H \pm 1\rangle$, four LH single exciton states $|L \pm 1\rangle$ and $|L \pm 0\rangle$, the HH and LH biexciton states $|HH0\rangle$ and $|LL0\rangle$, as well as four combined biexciton states $|HL \pm 2\rangle$ and $|HL \pm 1\rangle$. We label all these states by their valence band type and their total angular momentum;

Fig. 7.1 Calculated absorption spectrum for σ^- polarized light at zero magnetic field and as contour plot for increasing magnetic field.



in the case of the LH excitons with angular momentum zero the sign refers to the angular momentum of the hole. The Mn spin M has the quantum number $M = \frac{5}{2}$ and thus six possible orientations $M_z = \pm\frac{5}{2}, \pm\frac{3}{2}, \pm\frac{1}{2}$. With the six states of the Mn atom the full system comprises 90 states, labeled by their exciton state and their Mn spin states $|X; M_z\rangle$.

For this systems the Hamiltonian reads

$$\begin{aligned} \mathcal{H} = & \sum_X \epsilon^X |X\rangle \langle X| - (E^* \cdot P + E \cdot P^*) \\ & + (g_e \mu_B B \cdot S^e + g_h \mu_B B \cdot S^h + g_{Mn} \mu_B B \cdot M) \\ & + j_e (M \cdot S^e) + j_h (M \cdot S^h) + j_{eh} (S^e \cdot S^h). \end{aligned} \quad (7.1)$$

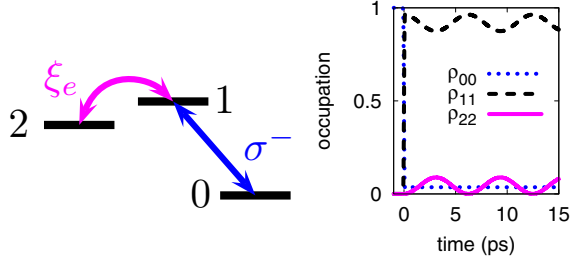
The first term in the Hamiltonian describes the energies ϵ^X of the excitons $|X\rangle$. The coupling of the carriers to an external light field E via the polarization operator P is treated in the usual dipole and rotating wave approximation in the second term. We will consider only Gaussian shaped laser pulses of the form $E = (E_0/\sqrt{4\pi\tau}) \exp[-(t-t_0)^2/\tau^2 - i\omega t]$ with the temporal width τ . The pulses have a width of $\tau = 85$ fs corresponding to a full width at half maximum of the intensity of 100 fs. A σ^+ (σ^-) circularly polarized pulse with pulse area π and resonant on the exciton transition creates a bright exciton with angular momentum $+1$ (-1). Single exciton states with a different value of the angular momentum thus describe dark excitons. The second line in Eq. (7.1) denotes the Zeeman coupling of an external magnetic field to electron, hole and Mn with the g -factors $g_e = -1.5$, $g_h = -0.1$, and $g_{Mn} = 2$, respectively. The third line describes the coupling of the carriers to the Mn spin via the exchange interaction as well as the electron-hole exchange interaction. Because the exchange is local, the carrier-Mn coupling strength strongly depends on the position of the Mn spin in the quantum dot. We take $j_h = 0.423$ meV and $j_e = -j_h/4$ which, assuming a confinement according to a square well potential [15] corresponds, e.g., to a dot with lengths of $L_z = 2$ nm and $L_x = L_y = 7$ nm, where the Mn atom is located approximately halfway between the dot center and the bor-

der. The electron-hole exchange coupling constant is taken to be $j_{eh} = -0.66$ meV. For the calculations the Liouville-von Neumann equation for the density matrix in the basis of the states $|X; M_z\rangle$ has been set up and solved numerically. Relaxation and dephasing processes caused by the radiative decay of the excitons have been included, but turned out to be of minor importance or even completely negligible [28]. Therefore we will not include this process here. From the optical response to a short and sufficiently weak laser pulse the absorption spectrum of the system can be calculated. Figure 7.1 shows the result for the case of excitation by σ^- polarized light in the region of the HH exciton transitions. In the left part we have plotted the spectrum at vanishing magnetic field while the right part shows the magnetic field dependence. We have assumed equal occupancies of the six Mn spin states. In agreement with luminescence spectra obtained both from the experiment [12] and from a numerical diagonalization of the Hamiltonian [15] we find at zero field six equally spaced lines which are shifted in energy with increasing magnetic field. Because the diamagnetic shift is not included in our model, this shift is linear with the field. In the field range between 7 T and 9 T all curves except the leftmost one exhibit an anticrossing which reflects the mixing of bright and dark excitons caused by the exchange interaction between electrons and Mn spin. We will come back to this point later. The most interesting part of the Hamiltonian for our purposes is the exchange interaction. We can rewrite it, e.g., for the electron-Mn spin coupling, as

$$M \cdot S^e = M_z S_z^e + \frac{1}{2} (M_+ S_-^e + M_- S_+^e), \quad (7.2)$$

where we have introduced the ladder operators $S_\pm^e = S_x^e \pm iS_y^e$ and $M_\pm = M_x \pm iM_y$. Analogous formulas hold for the other exchange interactions. Here, two different types of terms can be clearly distinguished. The first one is the Ising term and it describes an energy shift depending on the z -component of the spins. The second one is a flip-flop term, which describes simultaneous spin flips of electron and Mn spin, i.e., an electron can change its spin from up to down while the Mn increases its spin by one or vice versa. When looking in the spectral domain, this spin flip term gives rise to the coupling between bright and dark excitons and thus to the anticrossings seen in Fig. 7.1. In the time domain, on the other hand, it opens up the possibility to change the Mn spin state by an optical control of the exciton, since now the spin can be transferred from the exciton to the Mn ion. It should be noted that in a pure HH exciton system no correlated spin flip between hole and Mn spin is possible, because there is no coupling between the hole states with $S_z^h = +\frac{3}{2}$ and $S_z^h = -\frac{3}{2}$ introduced by the exchange Hamiltonian. On the other hand, the Ising term caused by the hole-Mn exchange interaction turns out to be mainly responsible for the splitting of the six lines observed in PL and absorption spectra because of the large coupling constant between Mn and hole.

Fig. 7.2 Left panel: Scheme of the three-level system including carrier-light interaction with a σ^- polarized pulse and exchange interaction via the coupling matrix element ξ_e . Right panel: Occupancies ρ_{ii} of the three involved states after excitation with a single pulse at $t = 0$.



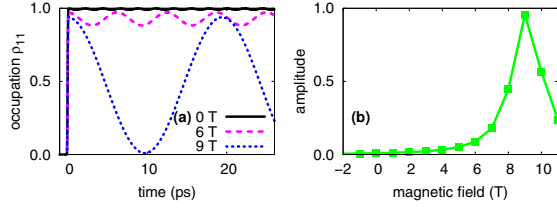
7.3 Spin flip in the heavy hole exciton system using π and 2π pulses

Let us first consider a single spin flip at a finite magnetic field of $B_z = 6$ T in the case of a pure HH exciton system. As mentioned above, because of $S_z^h = \pm \frac{3}{2}$ the hole spin is pinned and only the electron can induce spin flips via the exchange interaction [see Eq. (7.2)]. We assume that the Mn spin is initially in the state with $M_z = -\frac{5}{2}$ which, in the presence of a magnetic field, is indeed its ground state. We then apply a σ^- polarized π -pulse, which drives the system from the ground state $|0\rangle = |0; -\frac{5}{2}\rangle$ to the bright exciton state $|1\rangle = |H-1; -\frac{5}{2}\rangle$. Via the electron-Mn exchange interaction the state $|1\rangle$ couples to the dark state $|2\rangle = |H-2; -\frac{3}{2}\rangle$, which manifests itself in an oscillation of the occupancies of the latter two states. The occupancies of the three involved states ρ_{ii} are depicted in Fig. 7.2 after the excitation with a pulse at $t = 0$. The occupancies ρ_{11} rises from 0 to almost 1 during the laser pulse on a timescale of 100 fs, while simultaneously the occupancies of ρ_{00} drops to zero. Both occupancies ρ_{11} and ρ_{22} then start to oscillate with a period of about 6.5 ps.

Figure 7.2 suggests that the main dynamics of the whole system can be reduced to the dynamics in the three level system sketched in the left part of this Figure. The corresponding equations of motion for the density matrix within this three-level system read:

$$\begin{aligned}
 i\frac{d}{dt}\rho_{00} &= \Omega\rho_{01} - \Omega^*\rho_{01}^*, \\
 i\frac{d}{dt}\rho_{11} &= -(\Omega\rho_{01} - \Omega^*\rho_{01}^*) + \xi_e(\rho_{12} - \rho_{12}^*) \\
 i\frac{d}{dt}\rho_{22} &= -\xi_e(\rho_{12} - \rho_{12}^*), \\
 i\frac{d}{dt}\rho_{01} &= \frac{\mathcal{E}_1}{\hbar}\rho_{01} + (\Omega^*\rho_{00} - \Omega^*\rho_{11}) + \xi_e\rho_{02}, \\
 i\frac{d}{dt}\rho_{02} &= \frac{\mathcal{E}_2}{\hbar}\rho_{02} - \Omega^*\rho_{12} + \xi_e\rho_{01}, \\
 i\frac{d}{dt}\rho_{12} &= \frac{\mathcal{E}_2 - \mathcal{E}_1}{\hbar}\rho_{12} - \Omega\rho_{02} + \xi_e(\rho_{11} - \rho_{22}),
 \end{aligned} \tag{7.3}$$

Fig. 7.3 (a) Occupancy ρ_{11} as function of time for different magnetic fields B_z . (b) Amplitude of the oscillation as a function of the magnetic field B_z .



where ρ_{nn} refers to the occupancies of the state n and $\rho_{nn'} (n \neq n')$ to the coherence between the corresponding states. The coupling to the laser field is described by the matrix element $\Omega = d \cdot E / \hbar$ with the dipole matrix element d , and the exchange coupling matrix element is given by $\xi_e = j_e \sqrt{5} / (4\hbar)$. $\mathcal{E}_n$ are the energies of the respective states; the energy of the ground state is set to zero. Indeed it turns out that the dynamics obtained from this three-level model almost exactly reproduces the result shown in Fig. 7.2, which has been calculated within the full model including all 90 states.

As can be seen from Fig. 7.2, the interaction with the light field and the exchange interaction are acting on two very different time scales. This suggests to solve the equations of motion separately for these two processes. The light field coupling is only present during the laser pulse, i.e., on a 100 fs time scale. During the pulse we have $\Omega \gg \xi$. If the frequency of the light field is given by $\mathcal{E}_1 / \hbar$, the laser field drives resonantly the two level system formed by $|0\rangle$ and $|1\rangle$ in Eq. (7.3), thus creating or annihilating an exciton while the Mn spin is not affected. On the other hand, after the pulse we have $\xi \gg \Omega$; thus the exchange interaction is dominant coupling the states $|1\rangle$ and $|2\rangle$ and driving Rabi oscillations on the ps time scale by the constant exchange interaction. Since the states $|1\rangle$ and $|2\rangle$ are split by an energy difference $(\mathcal{E}_2 - \mathcal{E}_1)$ determined by the Ising terms of the three exchange interactions as well as by the corresponding Zeeman terms, these Rabi oscillations are off-resonant giving rise to a Rabi frequency $\omega_R = \sqrt{((\mathcal{E}_2 - \mathcal{E}_1) / \hbar)^2 + \xi_e^2}$ and to a reduced amplitude of the oscillations.

The frequency and amplitude of the Rabi oscillations depend strongly on the energy difference between the states $|1\rangle$ and $|2\rangle$. This energy difference can be efficiently manipulated by a magnetic field in Faraday configuration. Figure 7.3(a) shows the Rabi oscillation of the occupancy ρ_{11} after the single pulse excitation at $t = 0$ for three different magnetic fields $B_z = 0$ T, 6 T and 9 T. At zero magnetic field the energy splitting between dark and bright state is large and thus only a very weak oscillation can be seen. With increasing magnetic field the energy difference of the state becomes smaller and the amplitude of the Rabi oscillations increases. At a magnetic field of 6 T it is about 0.1, while at $B_z = 9$ T the energies of the states $|1\rangle$ and $|2\rangle$ are almost equal and thus the amplitude of the Rabi oscillation is close to one. The exciton system now oscillates completely between the bright and the dark state. A quantitative analysis of the amplitude as a function of the magnetic field is shown in Fig. 7.3(b). At $B_z = 9$ T there is the maximum of the amplitude. In the spectral domain the exchange coupling leads to the formation of new eigenstates

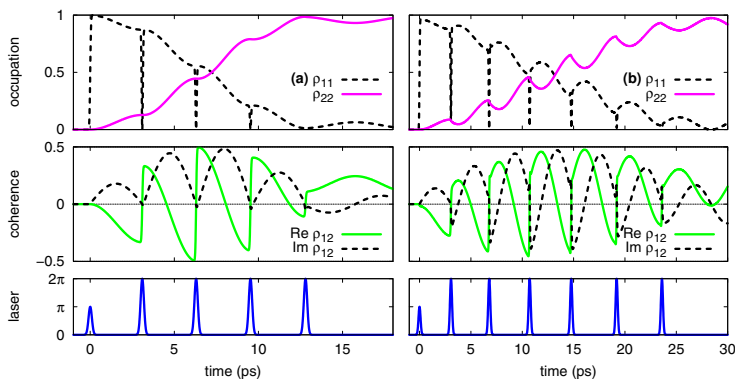


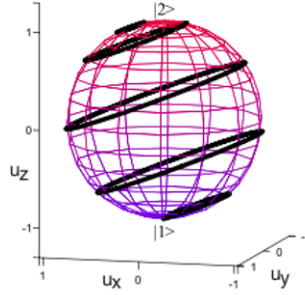
Fig. 7.4 Occupancies ρ_{11} and ρ_{22} (upper panel) as well as real and imaginary part of the coherence ρ_{12} (middle panel) for the case of excitation by a pulse sequence with one π and a series of 2π pulses (lower panel) (a) for an effective three-level system and (b) for the full model at a magnetic field of 6 T applied in Faraday configuration.

and to the anticrossings seen in the absorption spectrum of Fig. 7.1, where indeed the rightmost line exhibits an anticrossing at a magnetic field of 9 T.

From Fig. 7.3 it becomes clear that after a single pulse excitation a complete transfer from the electron spin to the Mn spin is only possible under resonance condition which, in our system, means at a magnetic field of 9 T. In this case, however, the Mn spin oscillates periodically between its values of $-\frac{5}{2}$ and $-\frac{3}{2}$. Therefore the question arises: Can we drive the Mn spin in such a way that it – at least approximately – afterwards remains in one of the other spin eigenstates. This is indeed possible by using additional pulses. For this purpose it is important to notice that even if we may treat the optical excitation and the exchange-induced dynamics separately because of the different time scales, they are not independent of each other. This is because the light field, which induces the dynamics in the effective two-level system consisting of the states $|0\rangle$ and $|1\rangle$, also acts on the coherences ρ_{02} and ρ_{12} connecting this subsystem with the third state $|2\rangle$. Especially when a 2π pulse is applied, this has an unexpected influence on the dynamics. A 2π pulse does not change the occupancy of the states $|0\rangle$ and $|1\rangle$, neither does it change the coherence between these states, but the coherences ρ_{02} and ρ_{12} are nevertheless affected. A 2π pulse introduces a global phase factor of -1 in the two-level system, which does not show up in density matrix elements within such a system, but it changes the sign of all coherences with states outside this two-level system. Thus, in our case the signs of the coherences ρ_{02} and ρ_{12} are changed. This in turn strongly influences the properties of the exchange-induced Rabi oscillation between the states $|1\rangle$ and $|2\rangle$. We can exploit this feature to drive the system from state $|1\rangle$ to state $|2\rangle$ with a series of 2π pulses at any value of the magnetic field.

To demonstrate such a switching process, Fig. 7.4(a) shows the occupancies ρ_{11} and ρ_{22} (upper panel) and coherence ρ_{12} (middle panel) obtained from an effective three-level model for the case of excitation by a π pulse and a subsequent series of

Fig. 7.5 Bloch sphere representation for the dynamics of the two-level model consisting of the states $|1\rangle$ to $|2\rangle$ for a sequence of 2π laser pulses resonant to the transition from $|0\rangle$ to $|1\rangle$. u_x , u_y , and u_z denote the three components of the Bloch vector as defined in the text.



2π pulses. All pulses have a σ^- polarization. The energies and coupling strengths of the three-level model have been taken from the full model at a magnetic field of 6 T. First of all one clearly notices that now a complete switching is achieved. The occupancy ρ_{11} goes to zero, while ρ_{22} goes up to one. If we look at the dynamics in detail, first ρ_{11} becomes occupied at $t = 0$ due to the π pulse. As before, both occupancies ρ_{11} and ρ_{22} start to oscillate. The first 2π pulse is applied at the minimum of the oscillations of the occupancy ρ_{11} . Due to the 2π pulse the occupancy ρ_{11} drops to zero, but immediately rises again to its previous value. Because the state $|2\rangle$ does not couple to the light field, its occupancy ρ_{22} is not affected by the laser pulse. However, the coherence ρ_{12} , which was also oscillating in time, is inverted. Due to this inversion of the off-diagonal element, the former minimum of the oscillation now becomes its maximum. This results in shifted boundaries for the exchange-induced Rabi oscillations. The frequency of the oscillation stays the same. To achieve a complete inversion of the occupancy we can repeat this process by using a series of 2π pulses, which are applied whenever the occupancy ρ_{11} reaches its minimum. In Fig. 7.4(a) it is clearly seen that each time a 2π pulse acts on the system, the coherence makes a phase jump and the occupancy oscillates between new boundaries. This eventually leads to the flip of the electron and Mn spin by one.

A very good visualization of the dynamics within the two-level system consisting of the states $|1\rangle$ and $|2\rangle$ is the representation in terms of the Bloch vector, which is defined by $u = (2\text{Re}(\rho_{12}), -2\text{Im}(\rho_{12}), 2\rho_{22} - 1)$. Since we consider no dissipation processes the length of the Bloch vector is fixed and its dynamics can be displayed on the Bloch sphere as shown in Fig. 7.5. If the Bloch vector points to the south (north) pole, the system is in the state $|1\rangle$ ($|2\rangle$). The off-resonant Rabi oscillations are represented by tilted circles on the sphere. Immediately after the first π pulse the Bloch vector points to the south pole. Then the exchange Rabi oscillation sets in which is represented by the bottom circle. When the occupancy ρ_{11} has reached its minimum, the Bloch vector is at the highest point of the bottom circle. If now a 2π pulse is applied, the x - and y -components of the Bloch vector are inverted; thus it jumps to the bottom of the next circle, which represents the dynamics of the new oscillation. With each 2π pulse the Bloch vector jumps to the next circle just as climbing up a ladder until it almost reaches the north pole, i.e., the system is in the states $|2\rangle$.

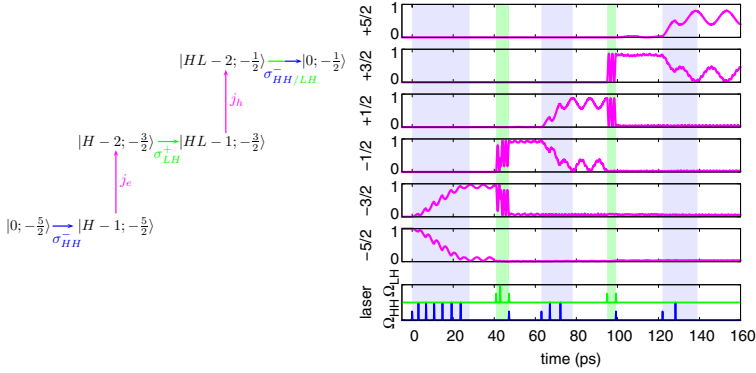


Fig. 7.6 Left panel: Scheme of the switching process via excitation, manipulation and annihilation of HH and LH excitons with circularly polarized laser pulses $\sigma^\pm$. Right panel: Occupancy of the six Mn spin eigenstates M_z as well as amplitudes $\Omega_{HH/LH}$ of the laser pulses tuned to the HH and LH transition, respectively, for a magnetic field of 6 T applied in Faraday configuration. The shaded areas indicate switching times, while in the white areas no pulses are applied.

The three-level model which has been used to explain and visualize the dynamics captures the essential physics of the process. When using the full model there are noticeable deviations from the ideal case, as is shown in Fig. 7.4(b). In particular the phase jumps are shifted due to the presence of the LHs and the electron-hole exchange interaction as well as the slightly off-resonant laser pulse. As a result, the minimum of ρ_{11} in one oscillation does not exactly become the maximum of the next oscillation. Nevertheless we find that a complete switching is still achieved.

7.4 Switching into all Mn spin states

When we want to switch into all Mn spin states, we need to expand our system, because after the flip of the electron spin and the change of the Mn spin by one, the exciton system is in a dark state, which is not optically addressable anymore by pulses tuned to the HH exciton frequency. After a single flip the Mn spin could only flip back to its initial value. To overcome this bottleneck, we include transitions to the LH excitons in our system. The LH excitons have a spin of $S_z^h = \pm \frac{1}{2}$, therefore they can perform spin flips and further change the Mn spin, such that we can now reach all six states of the Mn spin.

7.4.1 Switching into spin eigenstates

In order to switch into each of the six eigenstates M_z of the Mn spin we propose a switching protocol schematically shown in the left part of Fig. 7.6. By using this

scheme we can change the Mn spin by two via optical excitation, manipulation and annihilation of excitons. After the switching the excitons are removed from the system returning it to the excitonic ground state. Thus, angular momentum has been effectively transferred from the light field to the Mn spin. To further increase the Mn spin M_z this scheme can then be repeated. The right part of Fig. 7.6 shows the occupancy of the six Mn spin states under the excitation by the proposed laser pulse sequence with transitions of the HH and LH excitons again for the case of a magnetic field of 6 T applied in Faraday configuration. The corresponding pulse amplitudes Ω_{HH} and Ω_{LH} are displayed in the lower panel. To demonstrate that indeed a switching of the Mn spin into all states has been achieved we have included pulse-free periods (white areas) between the periods when the pulse sequences are applied (shaded areas). Starting from $M_z = -\frac{5}{2}$, we first use the same sequence of σ^- pulses on the HH transition shown in Fig. 7.4(b) turning the bright ($H-1$) exciton into the dark ($H-2$) exciton by a spin flip of the electron. Meanwhile the Mn goes from $M_z = -\frac{5}{2}$ to $M_z = -\frac{3}{2}$. Indeed the occupancy of $M_z = -\frac{5}{2}$ has almost vanished and $M_z = -\frac{3}{2}$ is almost fully occupied. After a short break time a σ^+ polarized pulse creates an additional LH exciton exciting the system to the combined ($HL-1$) biexciton. Because the conduction band is now completely filled and the HH spin is pinned, only the LH can perform a spin flip, which again rises the Mn spin. Because the splitting between bright and dark states in the light hole system is smaller compared to the HH system and the hole coupling is stronger than the electron coupling, this spin flip is much faster than the first flip, but is also off-resonant, as can be seen in Fig. 7.6. So we use one 2π pulse to achieve a complete flip. Now the electronic system is in the combined biexciton state ($HL-2$), which can be de-excited by two σ^- polarized π pulses, one of the HH transition and one of the LH transition. The exciton system is back in the ground state, i.e., no excitons are present in the system, but the Mn spin has changed by two. Indeed, in Fig. 7.6 the occupancy of $M_z = -\frac{1}{2}$ is close to one, while all other occupancy are almost zero. This state is now the new initial state for the proposed switching scheme to further change the Mn spin. As can be seen in Fig. 7.6, by a pulse sequence of only 3 pulses the occupancy of the Mn spin state $M_z = +\frac{1}{2}$ rises to one and then by a transition on the LH exciton $M_z = +\frac{3}{2}$ becomes occupied. When the Mn is in the state $M_z = +\frac{3}{2}$ again no exciton is present in the system and now the Mn spin has changed by four in total. The addressing of the final state $M_z = +\frac{5}{2}$ can be achieved with a pulse sequence on the HH exciton transition giving rise to the electron spin flip and ending in the dark HH exciton state ($H-2$). Thus, by this switching scheme all six Mn spin eigenstates have been addressed by optical means.

When looking at the occupancies of the Mn spin states in the pulse-free periods (white areas in Fig. 7.6) we notice that for $M_z = -\frac{5}{2}$, $-\frac{1}{2}$, and $+\frac{3}{2}$ the occupancies are almost constant while for $M_z = -\frac{3}{2}$, $+\frac{1}{2}$, and $+\frac{5}{2}$ increasingly pronounced oscillations are present. This is because in the former case no excitons are present in the system and thus all exchange terms are inactive, while in the latter case a HH exciton is still present giving rise to exchange-induced Rabi oscillations between bright and dark states. The increase of the oscillation amplitudes can be well understood

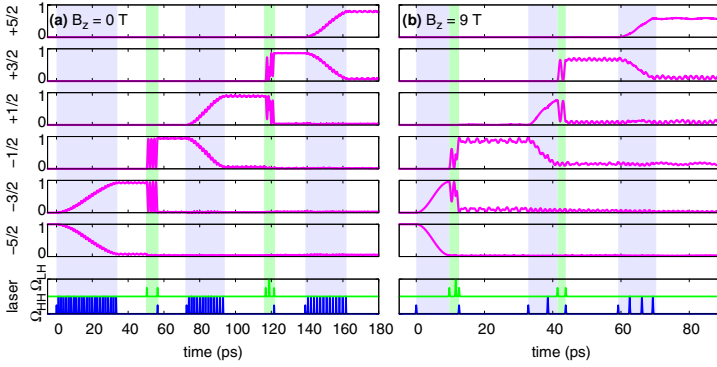


Fig. 7.7 Occupancies of the six Mn spin eigenstates M_z as well as amplitudes $\Omega_{HH/LH}$ of the laser pulses tuned to the HH and LH transition, respectively, (a) for the case without magnetic field and (b) for a magnetic field of 9 T. The shaded areas indicate switching times, while in the white areas no pulses are applied.

by looking at the absorption spectrum in Fig. 7.1. If we remember that the rightmost line corresponds to $M_z = -\frac{5}{2}$ and the leftmost line to $M_z = +\frac{5}{2}$ we clearly see that at a magnetic field of 6 T in our switching process we approach the anticrossing, i.e., the resonance between bright and dark exciton states. For the first switching step between $M_z = -\frac{5}{2}$ and $M_z = -\frac{3}{2}$ this resonance occurs at about 9 T. For the last switching step between $M_z = +\frac{3}{2}$ and $M_z = +\frac{5}{2}$ it occurs at about 7 T. Thus, at 6 T we are already rather close to this resonance, which explains the pronounced oscillations.

Because of its influence on the energy splittings between bright and dark exciton states, the magnetic field has strong impact on the total switching time and the number of pulses which are necessary. This can be seen in Fig. 7.7, where switching sequences are shown for zero magnetic field (left panel) as well as for $B_z = 9$ T. We notice that in both cases with the proposed switching scheme all Mn spin states can be addressed, but at zero magnetic field the number of pulses is very high, because of the large separation of the levels. On the other hand, in this case there are almost no oscillations in the occupancies of the Mn spin states even in the intervals where an exciton is present. On the contrary, at $B_z = 9$ T the first switching step is exactly in resonance, such that a single π pulse is sufficient; however, we cannot stop in the state with $M_z = -\frac{3}{2}$ because the Mn spin oscillates periodically between $M_z = -\frac{3}{2}$ and $M_z = -\frac{5}{2}$. Therefore the pulse on the LH exciton transition is applied immediately when $M_z = -\frac{3}{2}$ is reached. With increasing M_z the system moves away from the resonance. The occupancy of the final state $M_z = +\frac{5}{2}$ is almost constant even if there is a HH exciton present. These results show that the magnetic field is a very efficient control parameter, which can be used to optimize the switching sequence into a specific spin eigenstate.

The results discussed above have been obtained without taking into account any relaxation or dephasing processes. Spin relaxation processes are typically much

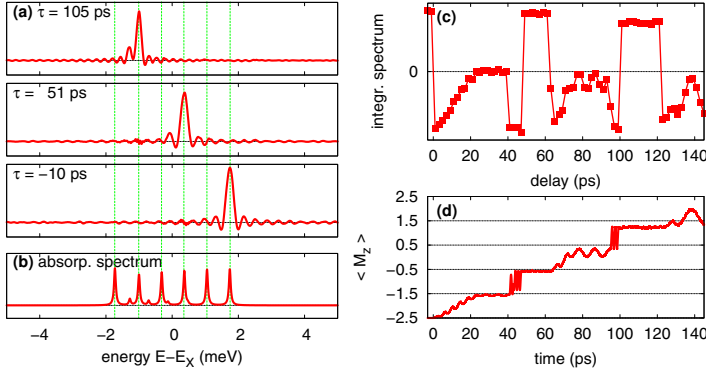


Fig. 7.8 (a) Probe spectra at various time delays τ for the switching sequence at $B_z = 6$ T as shown in Fig. 7.6. (b) Linear absorption spectrum at $B_z = 6$ T assuming equal occupancies of the six Mn spin states. (c) Integrated probe spectrum as function of time delay τ . (d) Expectation value of the Mn spin $\langle M_z \rangle$ as function of time t .

slower than the time scales considered here [26]. The radiative life time of the bright excitons, however, is often of the same order of magnitude as the total times shown in Figs. 7.6 and 7.7. Results of simulations where a radiative lifetime of 250 ps as measured in such systems [26] as well as the corresponding dephasing terms have been added to the Liouville-von Neumann equation in the standard way [30] can be found in Ref. [28]. These results show that there is only a small influence at low magnetic fields which becomes completely negligible at high magnetic fields. The reason is that even if, e.g. in Fig. 7.7(a), the total time of the switching process is almost 200 ps, bright excitons are present only for much shorter time intervals, and only during these intervals radiative processes can be active.

7.4.2 Measurement by pump probe spectroscopy

Having studied how the Mn spin state can be adjusted by optical means, we will now discuss how these spin dynamics can be measured optically. We already know that under steady state or equilibrium conditions the spin state M_z is reflected in the spectral position of the luminescence or absorption line of the exciton. The linear absorption spectrum for a magnetic field of $B_z = 6$ T and σ^- polarized light is shown in Fig. 7.8(b) under the assumption of equal occupancies of the six Mn spin eigenstates. The six lines correspond to transitions from the empty dot to a dot filled with one HH exciton with the Mn being in one of its six spin states from $M_z = +\frac{5}{2}$ to $M_z = -\frac{5}{2}$ from left to right. The smaller peaks appearing between the six main lines result from transitions from the empty dot to dark HH exciton states, which acquire a non-zero oscillator strength because of the coupling between bright and dark excitons caused by the the electron-Mn exchange interaction. The sensitivity of the

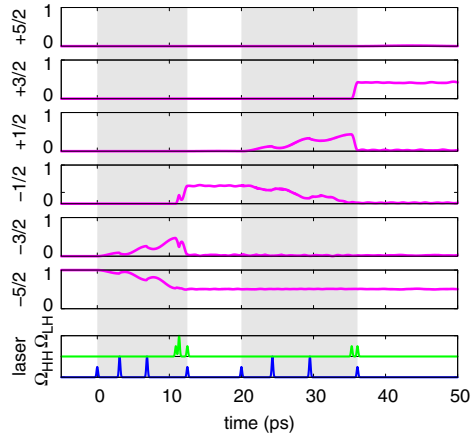
absorption spectrum to the Mn spin state suggests to use pump-probe spectroscopy to monitor its dynamics. In pump-probe experiments a pump pulse or a sequence of pump pulses with wave vector k_1 excites the system, while the response of the system to a probe pulse with wave vector k_2 is measured either in transmission or reflection geometry. To simulate such a pump-probe setup we take for the pump the same pulse sequence as above and we add a probe pulse at a given delay time τ with a variable phase $e^{i\phi}$ with respect to the first pulse. We then calculate the optical polarization as a function of this phase. This polarization $P(\phi, t)$ is expanded in a Fourier series according to

$$P(\phi, t) = \sum_n P_n(t) e^{in\phi}, \quad (7.4)$$

from which we obtain $P_1(t)$ as the desired probe polarization. The probe spectrum is then calculated in the usual way from the Fourier transform of the time-dependent polarization [31, 32]. As usual we take a probe pulse which is much weaker than the pump pulse, such that it does not much influence the dynamics of the system. With a duration of 10 fs it is also much shorter in time. To simplify the calculation of the spectrum we add a dephasing to the probe polarization with a dephasing time of 20 ps. Figure 7.8 shows absorption spectra of the probe pulse at three different delay times $\tau = -10$ ps, $\tau = 51$ ps and $\tau = 105$ ps for the switching dynamics of Fig. 7.6 at a magnetic field of 6 T. At all three delay times no exciton is in the system. Accordingly we see a peak in the absorption spectra of the probe pulse for all three delays, but at different energies. At $\tau = -10$ ps the system is in its initial state $M_z = -\frac{5}{2}$ and therefore we see only one peak at 1.7 meV, which equals the rightmost absorption peak. Now the switching takes place, which changes the Mn spin by two, and at $\tau = 51$ ps the Mn spin state $M_z = -\frac{1}{2}$ is fully occupied (Fig. 7.6). According to the new spin state of the Mn spin the energy of the single absorption peak has shifted to 0.3 meV. If we look at the last spectrum at $\tau = 105$ ps, a change of the Mn spin by four to $M_z = +\frac{3}{2}$ has occurred as we see in Fig. 7.6. Similar a single absorption peak comes up, which is now shifted to -1.0 meV. Since the whole pump laser sequence is applied, the probe polarization is also affected by subsequent pump pulses which gives rise to some spectral oscillations seen in all three spectra. But essentially all three spectra have a single absorption peak, and from the spectral position of this peak the Mn spin state can be extracted.

Another interesting feature of the dynamics is the occurrence of exchange-induced Rabi oscillations, which can be visualized by spectrally integrating the probe spectrum. Figure 7.8(c) shows the integrated probe spectrum for σ^- polarized light as a function of the delay time. Here the integration has been performed over the spectral region of the HH exciton transitions; thus, LH exciton contributions are not included. When no exciton is present in the system we expect the integrated spectrum to be positive corresponding to absorption. In contrast, if a bright HH exciton is present this leads to gain, which means that the integrated spectrum is negative. For comparison Fig. 7.8(d) shows the expectation value of the Mn spin state $\langle M_z \rangle$ for the switching sequence used in Fig. 7.6. We notice a clear correspondence between the temporal behavior of the spectrally integrated signal and this expecta-

Fig. 7.9 Occupancies of the Mn spin states M_z and applied laser sequence on the HH and LH exciton transition at a magnetic field of 6 T showing the creation of coherent superposition states of the Mn spin. Shaded areas mark switching processes and white areas indicate pulse-free intervals.



tion value. At negative delay times τ the integrated spectrum has a positive value because at negative times no exciton is present in the system. At $t = 0$ the bright exciton is created leading to a sign change in the integrated spectrum at $\tau = 0$. When the bright exciton is successively turned into a dark exciton, the integrated spectrum goes to zero, because when the dark exciton ($H - 2$) is present the optical transition to the bright exciton ($H - 1$) is blocked and the system becomes transparent. Indeed the integrated spectrum remains zero as long as $M_z = -\frac{3}{2}$. When in the further switching process the LH exciton is excited the integrated spectrum becomes negative again because now the HH can recombine with the newly generated electron. Then both excitons are removed from the system and the spectrum returns to absorption with positive values. This behavior is repeated in the second part of the switching sequence. Both in the intermediate period around 80 ps and at the final stage the signal also nicely reproduces the exchange-induced Rabi oscillations between bright and dark exciton states because in the former state there is gain while in the latter state the signal vanishes. We thus have found that pump-probe spectroscopy should indeed be a technique which is well suited to monitor the dynamics of the Mn spin.

7.4.3 Switching into superposition states

For applications in quantum information processing it is essential to be able to prepare coherent superposition states. Therefore it is interesting to study whether our switching protocol can be extended in such a way to generate also coherent superpositions of the Mn spin eigenstates. From the previous switching scheme we know that always after two steps the excitons can be removed from the system, which makes the Mn spin state particularly stable. Hence we seek for a switching protocol into a superposition of Mn spin states where M_z differs by two. Figure 7.9 shows an

example for such a pulse sequence by which a superposition between the $M_z = -\frac{5}{2}$ and either the $M_z = -\frac{1}{2}$ or the $M_z = +\frac{3}{2}$ state can be prepared. In the Figure we have again plotted the occupancies of the Mn spin states M_z as well as the laser pulse sequences on the HH and the LH exciton transitions at a magnetic field of 6 T. We again start with the Mn spin in the ground state $M_z = -\frac{5}{2}$ and σ^- pulses on the HH exciton transition to initiate the electron-Mn spin flip. But in order to achieve a superposition, when the occupancy of the $M_z = -\frac{3}{2}$ state reaches one half we already apply a σ^+ polarized pulse to inject a LH exciton. This has no effect on the state $|H-1; -\frac{5}{2}\rangle$, but changes the dark exciton state to the combined biexciton state $|HL-1; -\frac{5}{2}\rangle$ like before. In the latter state the conduction band is filled but the light hole can flip to $|HL-2; -\frac{1}{2}\rangle$. If we now apply two σ^- polarized π pulses, one resonant on the HH transition and the other resonant on the LH transition, both states are affected. The combined biexciton is destroyed to $|0; -\frac{1}{2}\rangle$, but the state $|H-1; -\frac{5}{2}\rangle$ changes to $|L-1; -\frac{5}{2}\rangle$. The latter state is stable, since all spins point into the same direction. We see in Fig. 7.9 that both occupancy $M_z = -\frac{5}{2}$ and $M_z = -\frac{1}{2}$ are one half. Indeed, the system is in a coherent superposition between the states $|L-1; -\frac{5}{2}\rangle$ and $|0; -\frac{1}{2}\rangle$. If we repeat the switching sequence starting from the previously generated superposition state with a σ^- polarized π pulse on the HH exciton transition, the state $|L-1; -\frac{5}{2}\rangle$ is transformed into $|HL-2; -\frac{5}{2}\rangle$ while the state $|0; -\frac{1}{2}\rangle$ changes into $|H-1; -\frac{1}{2}\rangle$. The former one is stable while the latter one starts to oscillate into the state $|H-2; +\frac{1}{2}\rangle$. Following the switching scheme of Fig. 7.6, we can drive this latter state into the state $|HL-2; +\frac{3}{2}\rangle$. Finally, two σ^- polarized π pulses on the HH transition and the LH transition remove the excitons from both contributions ending up with a coherent superposition between the states $|0; -\frac{5}{2}\rangle$ and $|0; +\frac{3}{2}\rangle$.

7.5 Magnetic field in Voigt configuration

A very different situation comes up when a magnetic field in Voigt configuration is present. Figure 7.10(a) shows the calculated absorption spectrum with σ^- polarized light in the presence of an increasing magnetic field B_x . The calculated spectrum is similar to the PL spectrum measured under these conditions [18]. In Voigt configuration the external magnetic field is perpendicular to the growth direction. Like in the case of the Faraday configuration the influence of the external field and of the exchange terms on the Mn spin can be combined in an effective magnetic field given by

$$B_{eff}^{Mn} = B + \frac{j_e}{g_{Mn}\mu_B} S^e + \frac{j_h}{g_{Mn}\mu_B} S^h. \quad (7.5)$$

However, while in Faraday configuration all contributions (on average) point in the z -direction, which therefore constitutes a fixed quantization axis, in Voigt configuration the three terms in general have different orientations: The external field points in x -direction, the exchange field of the heavy holes points in z -direction because

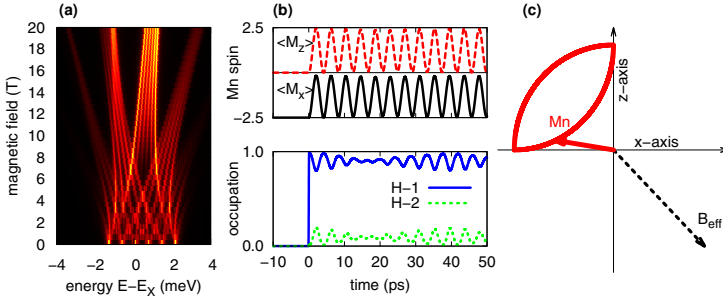
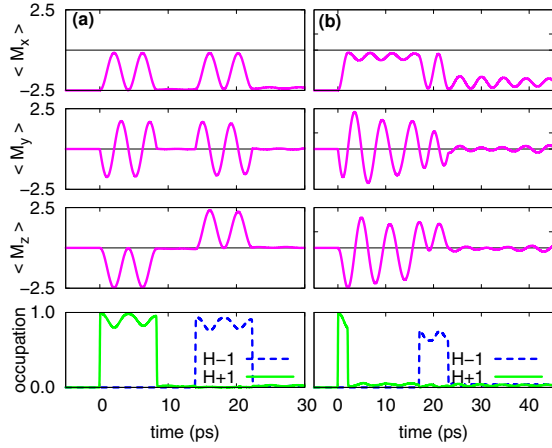


Fig. 7.10 (a) Calculated absorption spectrum for increasing magnetic field B_x in x -direction. (b) x - and z -component of the expectation value of the Mn spin $\langle M_{x,z} \rangle$ (upper panel) and occupancies of the bright exciton ($H-1$) and dark exciton ($H-2$) for the case of excitation by a single pulse on the HH exciton transition at a magnetic field of 6 T in Voigt configuration. (c) Scheme of the precession of the Mn spin around the effective magnetic field for an external field applied in Voigt configuration.

their angular momentum is determined by the growth direction and cannot flip, and the quantization axis of the electron is self-consistently determined by the effective magnetic field acting on the electron. This latter contribution is usually of minor importance, so we have essentially two contributions and the effective field lies in the xz -plane. At low magnetic fields the hole-Mn exchange term dominates and in the spectrum shown in Fig. 7.10(a) it gives rise to the six well-known equidistant lines. With increasing field each of the six lines splits and eventually the lines re-group forming a central line and a set of side lines, which are spaced by the Zeeman splitting of the Mn atom. A detailed explanation of the spectrum can be found in Ref. [18]. While the absorption spectrum reflects the eigenstates of the system let us now turn to the dynamical behavior after short-pulse excitation. Again we will study the case of an external magnetic field of 6 T, but now applied in the x -direction. In the ground state there is no exchange field. The effective field is oriented along the x -direction, which constitutes the quantization axis for the Mn spin. Thus, the ground state of the system is the state with $M_x = -\frac{5}{2}$. If by a circularly polarized laser pulse an exciton is created, the exchange contributions to the effective field are switched on and the axis of the magnetic field changes significantly from the x -axis to a direction in the xz -plane with, for our parameters, an angle of approximately 45° with respect to the x -axis as depicted in Fig. 7.10(c). Due to the change of the quantization axis the Mn spin starts to precess. For Mn doped quantum wells such a coherent precession of the Mn spins has been observed [33, 34] and also its coherent control has been demonstrated [35]. In these extended systems the precession is a collective effect which reflects an ensemble of many Mn atoms and which is usually well represented by treating the exchange interaction on the mean-field level. In contrast, here we are interested in the precession of a single Mn spin, i.e., we are dealing with a system that is too small to effectively build up a mean field.

Fig. 7.11 Cartesian components of the expectation value of the Mn spin $\langle M \rangle$ as well as occupancies of the exciton states $(H - 1)$ and $(H - 2)$ for two different pulse sequences in the presence of a magnetic field of 6 T applied in Voigt configuration.



Consequently, it can be expected that details of the time evolution of the electronic system are directly reflected in the dynamics of Mn spin.

For our parameters the precession period turns out to be $T = 4.2$ ps. The corresponding oscillations of the x - and z -components of the expectation values $\langle M_{x,z} \rangle$ are shown in Fig. 7.10(b). Since we start from $\langle M_x \rangle = -\frac{5}{2}$, the other components are zero before the pulse. A σ^- pulse at $t = 0$ changes the effective magnetic field and immediately the expectation values $\langle M_{x,y,z} \rangle$ start to oscillate. Because the angle between the effective magnetic field and the x -axis is 45° , the Mn spin rotates from $M_x = -\frac{5}{2}$, $M_z = 0$ to $M_x = 0$, $M_z = +\frac{5}{2}$ as indicated schematically in Fig. 7.10(c). Therefore, in Fig. 7.10(b) the amplitude of both oscillations is 2.5. The expectation value M_y oscillates symmetrically around zero (not shown). But also the electron is affected by the exchange field created by the Mn spin and the exchange field of the hole, which have different directions. Thus, also the electron spin starts to oscillate. However, as shown in the lower part of in Fig. 7.10(b) the amplitude of this oscillation is rather small because, the effective field is dominated by the electron-hole exchange interaction and therefore only slightly deviates from the z -direction.

With the creation and annihilation of excitons the axis of the effective magnetic field can be changed on the timescale of the laser pulse, which is much faster than the period of the exchange-induced Rabi oscillations. This gives the basis for an efficient control mechanism of the Mn spin dynamics also in Voigt configuration. In Fig. 7.11(a) we start again with the Mn spin $\langle M_x \rangle = -\frac{5}{2}$ and $\langle M_{y,z} \rangle = 0$ and apply a σ^+ polarized pulse to create the $(H + 1)$ exciton. As in the single pulse case, the Mn spin starts to precess. The z -component of the expectation value $\langle M_z \rangle$ oscillates between 0 and -2.5 reverse to the x -component $\langle M_x \rangle$, while the y -component of the expectation value $\langle M_y \rangle$ oscillates symmetrically around 0. After two periods of the oscillation we de-excite the exciton by another π pulse and thus switch off the exchange-induced component in the magnetic field. The effective field thus reduces to the external field. At this moment, the Mn already points along the magnetic field and therefore the precession ends and the Mn spin is stable again with $\langle M_x \rangle = -\frac{5}{2}$.

and $\langle M_{y,z} \rangle = 0$. If we now apply a σ^- polarized pulse and create a $(H - 1)$ exciton, the effective magnetic field points in the negative direction with respect to the z -axes. Thus again a precession of the Mn spin starts, but now $\langle M_z \rangle$ changes its sign and oscillates between 0 and $+2.5$. Also this oscillation can be switched off, when the exciton is annihilated after 2 periods.

The oscillation can also be controlled, if the exciton is annihilated when the Mn spin is pointing along the z -axis, i.e., $\langle M_z \rangle = -2.5$ as shown in Fig. 7.11(b). Now the Mn spin stands nearly perpendicular to the magnetic field, which without exciton is parallel to the x -axis, such that here $\langle M_z \rangle$ does a full rotation from $\langle M_z \rangle = -2.5$ to $\langle M_z \rangle = +2.5$ while $\langle M_x \rangle$ remains almost zero. When $\langle M_z \rangle$ is close to zero we excite the $(H - 1)$ exciton and change the axis of the effective magnetic field such that $\langle M_z \rangle$ is restricted to positive values and $\langle M_x \rangle$ starts to oscillate again. When $\langle M_x \rangle = -2.5$ is reached, we annihilate the exciton to return to our initial state. As one can see in Fig. 7.11(b) this switching is not perfect. Small oscillations are always present in the system due to the oscillations of the electron spin. Nevertheless, the optical control of the exciton provides a powerful tool for changing the effective magnetic field acting on the Mn spin and its precession dynamics.

7.6 Conclusions

In conclusion, we have studied coherent control mechanisms for a single quantum dot doped with a single Mn atom. By means of optical creation, manipulation and annihilation of excitons in the system it is possible to switch into all eigenstates of the Mn spin on a picosecond time scale, as well as to create coherent superpositions between these states. A magnetic field in Faraday configuration acts as a control parameter which can be used to optimize the switching dynamics with respect to switching time, required number of pulses and stability of the final state. With a magnetic field applied in Voigt configuration the precession of the Mn spin can be controlled via changing the effective magnetic field on the timescale of the laser pulses.

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Chapter 8

Coherent magneto-optical activity in a single chiral carbon nanotube

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Abstract We propose a theoretical framework and dynamical model for description of the natural optical activity and Faraday rotation in an individual chiral single-walled carbon nanotube in the highly nonlinear coherent regime. The model is based on a discrete-level representation of the optically active states near the band edge. Chirality is modeled by a system Hamiltonian in a four-level basis corresponding to energy-level configurations, specific for each handedness, that are mirror reflections of each other. The axial magnetic field is introduced through the Aharonov-Bohm and Zeeman energy-level shifts. The time evolution of the quantum system, describing a single nanotube with defined chirality, under an ultrashort polarised pulse excitation is studied using the coupled coherent vector Maxwell-pseudospin equations [Ref.[34]]. We provide an estimate for the dielectric response function and the optical dipole matrix element for transitions excited by circularly polarised light in a single nanotube and calculate the magnitude of the circular dichroism and the specific rotatory power in the absence and in the presence of an axial magnetic field. Giant natural gyrotropy (polarisation rotatory power $\sim 3000^\circ/\text{mm}$ ($B = 0$)), superior to the one of the crystal birefringent materials, liquid crystals and comparable, or exceeding the one of the artificially made helical photonic structures, is numerically demonstrated for the specific case of a (5,4) nanotube. A quantitative estimate of the coherent nonlinear magneto-chiral optical effect in an axial magnetic field is given ($\sim 30000^\circ/\text{mm}$ at $B = 8\text{ T}$). The model provides a framework for investigation of chirality and magnetic field dependence of the ultrafast nonlinear optical response of a single carbon nanotube.

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8.1 Introduction

The interaction of the polarised light with chiral materials in the absence of magnetic fields gives rise to the phenomenon optical (rotation) activity whereby the polarisation plane is rotated continuously during the light propagation across the medium. A simple phenomenological description of this effect, without considering the actual mechanisms involved, proposed by Fresnel, is based on the circular double refraction, or the difference of the phase velocity of propagation for the left- and right-circularly polarised light components. This in turn leads to a difference in the refractive indices, and/or absorption for left- and right-circularly polarised light which manifests itself as a circular birefringence, dichroism and rotatory power. Although the phenomenon of optical activity can be treated in terms of the classical electromagnetic theory, its in-depth understanding and full description requires a quantum-mechanical treatment. The early molecular models were of two classes: two-particle models, e.g. the models of Born, Oseen [2], Kuhn [3], Kirkwood [4] based on a spatial distribution of coupled oscillators, and single-oscillator models, e.g. Drude helix model [5] and Condon, Altar and Eyring theory [7] (for a comprehensive review consult [1]). The first consistent quantum mechanical theoretical formalism was put forward by Rosenfeld without invoking the couple oscillator model [6]. However after Kuhn demonstrated that, when correctly treated, the Drude single-oscillator model does not exhibit rotatory power, Drude model was long forgotten. It has been long thought that the notion of coupled oscillators is a necessary condition for the explanation of the rotatory power, until Condon, Altar and Eyring model [7] demonstrated optical activity using a quantum mechanical single-oscillator model. In the seventies, however, it has been shown [8] that Drude model exhibits optical activity in the nonlinear regime and recently it has been demonstrated that the model leads to optical activity if the motion of the particle is treated quantum mechanically [9].

The natural optical activity in chiral media, considered above, and the Faraday effect of magnetically induced optical activity, both manifest themselves as a rotation of the polarisation of the transmitted light. However the two effects are fundamentally different: while the natural optical activity is a result of a nonlocal optical response of a medium lacking mirror symmetry, the magnetic optical activity results from the breaking of the time-reversal symmetry by a magnetic field. The apparent similarity between the two effects has led to the theoretical prediction and experimental demonstration [26] of the link between the two phenomena through the so called magneto-chiral optical effect which occurs under conditions when both symmetries are broken simultaneously. Therefore formulation of a theory and model of the optical activity in chiral molecules, such as individual single-wall carbon nanotubes (SWCNTs), in the high-intensity nonlinear coherent regime and under an axial magnetic field, which is the subject of this paper, is of special interest from a fundamental point of view. Up to our knowledge no such theory has been proposed and very little is known about the polarisation dynamics of the nonlinear optical and magneto-optical response of a single carbon nanotube. Detailed understanding of the mechanisms underlying the optical and magneto-optical birefringence, circular

dichroism and rotation in the nonlinear coherent regime open up pathways for development of a novel class of ultrafast polarisation-sensitive optoelectronic devices, e.g. ultrafast optical switches, polarisers etc., based on single carbon nanotubes (CNTs) as basic functional components of an integrated optoelectronic device.

In the quantum theory of the optical activity one may distinguish two distinct classes of optically active molecules: a few molecules are inherently asymmetric, and the majority of the molecules owe their activity to the asymmetric juxtaposition of symmetric (inherently optically inactive) subsystems. Both mechanisms should be considered in a full treatment of the molecular optical activity model [10]. In what follows we shall consider single objects rather than ensembles of carbon nanotubes and the chiral symmetry results from the inherent spiral alignment of the C-atoms in a single SWCNT. Chirality is one of the main symmetries of the carbon nanotube geometry which determines the optical properties of the SWCNTs [12]. Investigation of the magneto-optical phenomena in chiral nanotubes is of considerable interest for the nonlinear optical spectroscopy since it provides important information about their electronic structure [11], [13, 14, 15, 16].

Magnetic circular dichroism (MCD) and magneto-optical rotatory dispersion (MORD) techniques offer spectroscopic information which is different or impossible to obtain by other means. Magneto-optic Faraday effect and the time-resolved Faraday rotation technique in particular [19], allows measurement of the optically-driven spin dynamics in semiconductor nanostructures on an ultrafast time scale and with high spatial resolution. The study of the spin dynamics in single carbon nanotubes is of particular interest in view of potential integration of carbon nanotubes in future spin devices.

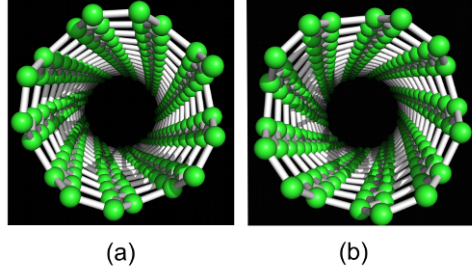
Another motivation for the present study is that the chiral materials represent interest on their own as they exhibit negative refractive index in a given frequency range and are promising candidates for metamaterials applications. Metamaterials consisting of helical structures have been long recognised as promising candidates in the microwave range, however for applications working in the visible range, the helical structures should be down-scaled to nanometer size and therefore natural candidates could be sought at a molecular level. In fact, it has been demonstrated that some helical molecules show strong optical activity [20, 21, 22, 23, 24, 25]. Recently giant optical gyrotropy in the visible and near-IR spectral ranges, of up to $2500^\circ/\text{mm}$ has been demonstrated in artificial gyrotropic chiral media, in view of implementations as a photonic chiral negative index metamaterial [27]. In this respect, it would be of great interest to assess the magnitude of the natural and Faraday optical rotation in single carbon nanotubes as 1D-helical molecules under resonant coherent optical excitation, and to compare it with the one of other candidates for metamaterials working in the visible range. A theoretical framework for discussion of the optical activity [17] and Faraday rotation phenomena [18] in CNTs has been proposed within the tight-binding formalism. However, up to our knowledge, the way the chirality affects the ultrafast nonlinear optical response of a single nanotube and the magnitude of this effect without and in an external magnetic field, has not been investigated. In this paper we develop a dynamical model for the ultrafast circularly-polarised light pulse interaction with single chiral SWC-

NTs in the absence and in the presence of an external magnetic field which allows for assessing the magnitude and ellipticity of this rotation. The chapter is organised as follows. In Sec. 8.2 we formulate the problem of ultrashort circularly-polarised pulse propagation and resonant coherent interactions in an isolated SWCNT. The proposed dynamical model for description of the ultrafast optically-induced dynamics requires both the knowledge of the dielectric response function and the optical dipole matrix element for optical transitions excited by circularly polarised light in a single SWCNT. The calculation of these model parameters is given in respective subsections of Sec. 8.2. In Sec. 8.3 we provide a theoretical framework for tackling the resonant nonlinear absorption/amplification in chiral carbon nanotubes based on a band-edge energy level configurations specific for left- and right- circularly polarised optical excitation. In Sec. 8.4 we present the simulation results for the ultrafast nonlinear dynamics of the natural optical activity in a single chiral (5,4) SWCNT and calculate the time- and spatially resolved circular dichroism, birefringence and specific rotatory power for a particular single nanotube geometry. In Sec. 8.5 we develop a model of the nonlinear Faraday rotation in an axial magnetic field in a chiral SWCNT, including both the energy level shift due to the Aharonov-Bohm flux and the spin-B interaction resulting in Zeeman splitting of the energy levels near the band gap. We provide an estimate for the spatially resolved circular dichroism and specific rotatory power along the tube length following the initial pulse excitation. Finally, in Sec. 8.6 we give a summary of the results and outline some future model applications.

8.2 Problem Formulation

SWCNTs are uniquely determined by the chiral vector, or equivalently by a pair of integer numbers (n,m) in the planar graphene hexagonal lattice unit vector basis. A primary classification of carbon nanotubes is the presence or the absence of the chiral symmetry. Achiral nanotubes, whose mirror image is superimposable, are subdivided into two classes: zig-zag ($m=0$) and armchair ($m=n$) nanotubes. The rest of the nanotubes belong to the most general class of chiral nanotubes, whose mirror reflection is not superimposable. Chiral molecules exist in two forms that are mirror images of each other (enantiomers). Similarly, carbon nanotubes exist in two (AL) left- and (AR) right-handed helical forms depending on the rotation of two of the three armchair chains of carbon atoms counterclockwise or clockwise when looking against the nanotube z -axis. The two helical forms are shown for illustration for a (5,4) and (4,5) SWCNT in Fig. 8.1, where we have adopted the following convention: $m > n$ corresponds to a left-handed (AL) nanotube, whereas $m < n$ corresponds to a right-handed (AR) nanotube. The electronic band structure of a SWCNT [31] is described by the quantization of the wavevector along the tube circumference, perpendicular to the tube axis resulting in a discrete spectrum of allowed k -vector states forming subbands in the valence and conduction band labeled by the allowed quasi-angular momentum number (μ). When a circularly

Fig. 8.1 An illustration of the single carbon nanotube molecular structure of (a) left-handed AL (5,4) and (b) right-handed AR (4,5) single SWCNT calculated using the tight-binding method of P_z -orbitals [30]: a view along the tube axis against z-axis direction



polarised (in the x-y plane) laser pulse propagates along the z-axis of an AL or AR SWCNT (Fig. 8.2), only one of the two allowed transitions for a linearly polarised light (along x or y), between the quasi-angular momentum sub-band states $\mu \rightarrow \mu - 1$ and $\mu \rightarrow \mu + 1$, can be excited [29](Fig. 8.2). Here we adopt the following convention for the optical pulse polarisation: the left ($\sigma^- = x - iy$) and right ($\sigma^+ = x + iy$) helicity of light corresponds to counterclockwise and clockwise rotation of the electric field polarisation vector when looking towards the light source (against z-axis direction).

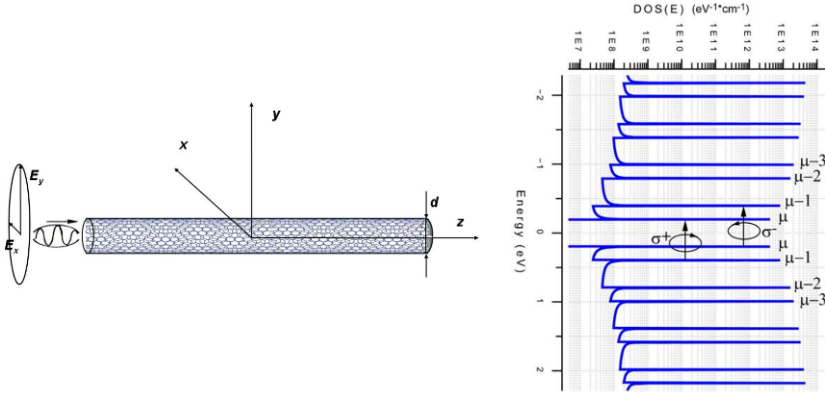


Fig. 8.2 Left, geometry of an experiment with an optical excitation by circularly polarised pulse propagating along the SWCNT axis; Right, 1D electronic density of states (DOS) vs energy at the K-point of the Brillouin zone ($\mu > 0$) of a AL-handed (20, 10) chiral SWCNT. The allowed dipole optical transitions for circularly polarised light are designated by arrows for left- (σ^-) and right- (σ^+) pulse helicity.

We model the single chiral nanotube band-edge structure at the K point of the Brillouin zone by an ensemble of identical four-level systems, corresponding to the dipole optically allowed transitions for AL and AR nanotube enantiomers. As has been pointed out in [29], taking into account the polarisation sense convention above, absorption of right circularly polarised light σ^+ excites the electronic transitions $\mu - 1 \rightarrow \mu$, or equivalently $\mu \rightarrow \mu + 1$ in AL-handed SWCNTs and $\mu \rightarrow \mu - 1$ transitions for AR-handed SWCNTs, the absorption of left circularly polarised σ^-

light excites $\mu \rightarrow \mu - 1$ transitions for AL-handed and $\mu \rightarrow \mu + 1$ transitions for AR-handed SWCNTs (Fig. 8.3).

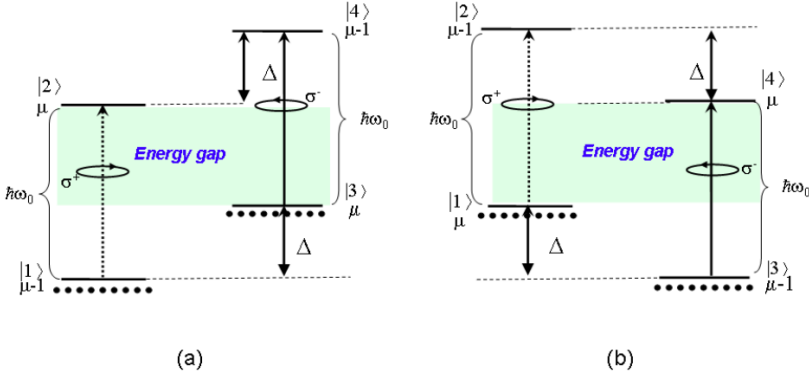


Fig. 8.3 Energy-level structure at the $K(K')$ point of the lowest subbands labeled by the subband index μ for an (a) AL-handed; (b) AR-handed SWCNT. The fundamental energy gap is shaded. ω_0 is the resonant transition frequency and Δ is the energy separation between the lowest subband and the second lowest subband near the band gap. The dipole optical transitions excited by σ^- and σ^+ circularly polarised light are designated by arrows. Only one of the two transitions is allowed for circularly polarised light, denoted by solid (dashed) arrow. Valence band states below the band gap are populated.

Note that the chiral symmetry of the medium is incorporated at a microscopic level through the energy level scheme and the allowed dipole optical transitions for AL- and AR- handed SWCNTs: the mirror reflections of the two energy-level configurations cannot be superimposed. The origin of the optical activity is the difference in the optical selection rules for left- and right-circularly polarised light and the specific relaxation channels involved at optical excitations with each light helicity with their respective timescales [33].

The proposed theoretical model, details of which will be given in Sec. 8.3, is based on the self-consistent solution of Maxwell's equations in vectorial form for the polarised optical pulse propagation and the time-evolution master pseudospin equations of the discrete multi-level quantum system in the external time-dependent perturbation [34]. This is a semiclassical approach which treats the optical wave propagation classically through the Maxwell's equations and therefore requires the knowledge of the effective dielectric constant of the medium. On the other hand, the quantum evolution equations require the knowledge of the dipole optical transition matrix element for transitions excited by circularly polarised light in a single carbon nanotube. In what follows we shall give an estimate of these important model parameters. Without loss of generality, we shall consider the specific (5,4) chiral AL-handed SWCNT which we shall assume to be $L_n = 500$ nm long. Tight-binding calculations [30] provide the electronic band structure (Fig. 8.4), the fundamental band gap $E_g = E_{\mu,\mu} = 1.321$ eV, corresponding to a wavelength $\lambda = 939$ nm, nan-

otube diameter $d = 0.61145 \text{ nm}$ and chiral angle $\theta = 26.33^\circ$. The resonant transition

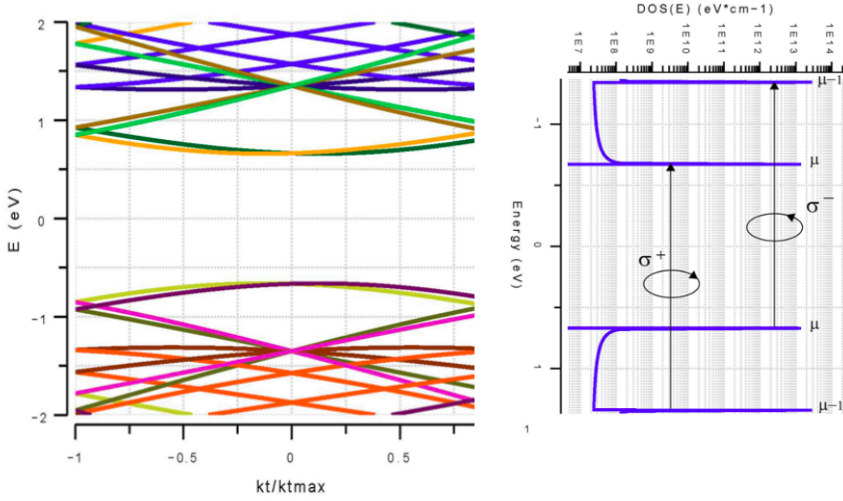


Fig. 8.4 Left, energy dispersion vs transverse (along the tube circumference) wavevector k_t , normalised with respect to the wavevector at the boundary of the Brillouin zone k_{tmax} of a (5,4) SWCNT; Right, 1D-Density of states vs energy showing the lowest energy subbands near the band edge involved in dipole optical transitions induced by circularly polarised light. Only one transition can be excited at any one time by each helicity (σ^- or σ^+)

energy for circularly polarised excitations is $E_{\mu, \mu \pm 1} = 1.9815 \text{ eV}$, corresponding to a resonant wavelength $\lambda_0 = 626.5 \text{ nm}$.

8.2.1 Dielectric response function of an isolated SWCNT

Our calculation of the dielectric constant of a single carbon nanotube is based on the effective medium approximation. Following the approach described in [37] the local dielectric tensor of a cylindrical carbon nanotube can be written in cylindrical coordinates as:

$$\epsilon(\hat{r}, \hat{\phi}, \hat{z}) = \epsilon_{\parallel} \hat{r} \hat{r} + \epsilon_{\perp} (\hat{z} \hat{z} + \hat{\phi} \hat{\phi}) \quad (8.1)$$

where $\epsilon_{\parallel}$ ($\epsilon_{\perp}$) are the principal components of the dielectric tensor of graphite parallel (perpendicular) to the normal axis of the graphite planes. The dielectric polarizability of a single carbon nanotube in an external potential of the form $V_m(r, \Phi) = V(r)e^{im\Phi}$, i.e. no field applied parallel to the tube z-axis which corresponds to the considered case of a circularly-polarised electromagnetic wave in a plane, perpendicular to the tube axis, an expression for the polarizability per unit length is given in [38] in terms of the inner, r and outer, R radii of the nanotube,

described as a hollow cylinder. In the quasistatic approximation $m = 1$ the nanotube polarizability is anisotropic, given by:

$$\alpha_1 = 4\pi\epsilon_0\epsilon_e R^2 \frac{(\epsilon_{||}\lambda - \epsilon_i)(\epsilon_{||}\lambda + \epsilon_e)\rho^{2\lambda} - (\epsilon_{||}\lambda - \epsilon_e)(\epsilon_{||}\lambda + \epsilon_i)}{(\epsilon_{||}\lambda - \epsilon_i)(\epsilon_{||}\lambda - \epsilon_e)\rho^{2\lambda} - (\epsilon_{||}\lambda + \epsilon_e)(\epsilon_{||}\lambda + \epsilon_i)} \quad (8.2)$$

where $\rho = r/R$, $\lambda = (\epsilon_{\perp}/\epsilon_{||})^{1/2}$, ϵ_i and ϵ_e are the dielectric constants of the internal and external materials, for a free standing nanotube, we shall assume $\epsilon_i = \epsilon_e = 1$.

An equivalent isotropic dielectric function, ϵ of a solid cylinder with the nanotube's external radius, R is introduced, according to:

$$\alpha'_1 = 4\pi\epsilon_0\epsilon_e R^2 \frac{\epsilon - \epsilon_e}{\epsilon + \epsilon_e} \quad (8.3)$$

The principal axes of the dielectric function tensor, $\epsilon_{||}$ and $\epsilon_{\perp}$ in Eq.(8.2) can be calculated from graphite ordinary and extraordinary ray refractive indices, $n_o = 2.64$ and $n_e = 2.03$, respectively [40]. Taking the inner radius $d = 0.61145$ nm of the (5,4) nanotube under consideration and a typical value for the external nanotube radius of $R \sim 5$ nm [39], and equalizing Eq.(8.2) and Eq.(8.3), one can obtain the equivalent isotropic dielectric constant of the anisotropic nanotube, $\epsilon = 5.32821$, giving a value $n \approx 2.3$ for the refractive index.

8.2.2 Optical dipole matrix element for circularly polarised light

For the calculation of the optical dipole matrix element under resonant circularly polarised pulse excitation, we assume injecting a resonant ultrashort circularly-polarized pulse with pulse duration $\tau = 60$ fs and excitation fluence $S = 20$ mJ/m², in agreement with the experiment conducted in [33, 36]. In order to get an estimate of the dipole matrix element, we employ the extension of the effective mass method applied to chirality effects in carbon nanotubes proposed in [35]. It has been shown that chirality effects can be described by an effective Hamiltonian, in which the coupling between the electron wavevector k_z along the tube axis and the quasangular momentum around the tube circumference is taken into account. Using this Hamiltonian a general expression for the electron-photon matrix element between the initial state i and final state f has been derived ([35], Eq. (18)) depending on the incident pulse light intensity I . In order to obtain a value for I , we calculate first the electric field amplitude corresponding to a power of S/τ , using the expression for the intensity of a plane wave P in [W/m²] in a dielectric [41]:

$$E = \sqrt{\frac{2P}{cn_{\text{refr}}\epsilon_0}} \quad (8.4)$$

where n_{refr} is the medium refractive index. The volume energy density of an electromagnetic wave in $[J/m^3]$ in the expression for the matrix element is then given by:

$$I = \frac{1}{2} \epsilon_0 n_{refr}^2 E^2 \quad (8.5)$$

Substituting the chiral indices $n = 5, m = 4$ and all parameters, e.g. the resonant energy for the transition induced by circularly polarised light, tube diameter, refractive index (see subsection A) in [35], Eq. (18) yields a dipole moment of $\wp = 3.613 \times 10^{-29} \text{ Cm}$. On the other hand, we could obtain an estimate of the dipole moment from the spontaneous emission rate of the $\mu \rightarrow \mu$ transition excited by linearly polarised light. Radiative lifetimes in the range $\tau_{sp} = 7 - 110 \text{ ns}$, comparable to those of the allowed molecular transitions, have been reported [36]. The corresponding dipole moments vary in the interval $\wp = 4.277 \times 10^{-29} - 1.079 \times 10^{-29} \text{ Cm}$, and the most likely radiative lifetime of 10 ns implies $\wp = 3.579 \times 10^{-29} \text{ Cm}$, which is in a very good agreement with the value calculated above using the extension to the effective mass method [35].

8.3 Theoretical framework for the natural optical activity

Consider an ultrashort circularly polarised pulse in the $(x - y)$ plane propagating along the nanotube z -axis, resonantly coupled to an ensemble of identical homogeneously broadened four-level systems describing the resonant absorption or amplification in a single carbon nanotube (Fig. 8.5). Following the coherent Maxwell-pseudospin formalism developed in [34], the generic system Hamiltonian in the four-level basis that applies to an excitation with either σ^- or σ^+ light helicity, is written at resonance, as follows:

$$\hat{H} = \hbar \begin{pmatrix} 0 & -\frac{1}{2}(\Omega_x + i\Omega_y) & 0 & 0 \\ -\frac{1}{2}(\Omega_x - i\Omega_y) & \omega_0 & 0 & 0 \\ 0 & 0 & \Delta & -\frac{1}{2}(\Omega_x - i\Omega_y) \\ 0 & 0 & -\frac{1}{2}(\Omega_x + i\Omega_y) & \Delta + \omega_0 \end{pmatrix} \quad (8.6)$$

where $\Omega_x = \wp E_x / \hbar$, $\Omega_y = \wp E_y / \hbar$ are the time-dependent Rabi-frequencies associated with E_x and E_y electric field components and $\wp$ is the optical dipole matrix element for $\mu \rightarrow \mu \pm 1$ transitions excited by circularly polarised light. The time evolution of the quantum system under an external dipole-coupling perturbation in the presence of the relaxation system (Fig. 8.5) is governed by a set of pseudospin master equations for the real state 15-dimensional vector S derived in [34]:

$$\frac{\partial S_j}{\partial t} = \begin{cases} f_{jkl} \gamma_k S_l + \frac{1}{2} \text{Tr} \left(\hat{\sigma} \hat{\lambda}_j \right) - \frac{1}{T_j} (S_j - S_{je}), & j = 1, 2, \dots, 12 \\ f_{jkl} \gamma_k S_l + \frac{1}{2} \text{Tr} \left(\hat{\sigma} \hat{\lambda}_j \right) & j = 13, 14, 15 \end{cases} \quad (8.7)$$

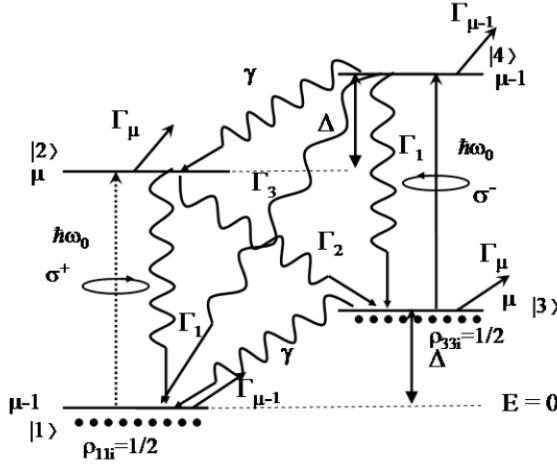


Fig. 8.5 Energy-level diagram in the general case of a resonant optical excitation of $\mu \rightarrow \mu + 1$ and $\mu \rightarrow \mu - 1$ interband transitions by σ^+ and σ^- polarised pulse, respectively. The resonant transition energy is $\hbar\omega_0$. Δ is the energy separation between the first and the second lowest conduction (valence) band states. The initial population of the lowest valence states below the band gap, ρ_{11i} and ρ_{33i} , is equally distributed between levels $|1\rangle$ and $|3\rangle$. All allowed longitudinal relaxation processes between the levels, associated with population transfer, are designated by wavy lines. Γ_1 is the spontaneous emission (radiative decay) rate of the interband transition $|2\rangle \rightarrow |1\rangle$, which owing to the symmetry is assumed equal to the spontaneous emission rate of $|4\rangle \rightarrow |3\rangle$; Γ_2 is the spontaneous emission rate for the $\mu \rightarrow \mu$ transition allowed for linearly polarised light; Γ_3 is the decay rate of the $\mu - 1 \rightarrow \mu - 1$ linearly polarised transition and γ is the intraband relaxation rate. The transverse relaxation (dephasing) rates Γ_μ and $\Gamma_{\mu-1}$ are designated by arrows.

where γ is the torque vector, f is the fully antisymmetric tensor of the structure constants of SU(4) group, and T_j are the phenomenologically introduced non-uniform spin decoherence times describing the relaxation of the real state vector components S_j toward their equilibrium values S_{je} . Using the generators of the SU(4) Lie algebra, the following expression for the torque vector is obtained:

$$\gamma = \left(-\Omega_x, 0, 0, 0, 0, -\Omega_x, \Omega_y, 0, 0, 0, 0, -\Omega_y, \omega_0, \frac{2\Delta - \omega_0}{\sqrt{3}}, \sqrt{\frac{2}{3}}(\Delta + \omega_0) \right) \quad (8.8)$$

In Eq.(8.7), similar to [42], we have introduced a diagonal matrix $\hat{\sigma} = \text{diag}(\text{Tr}(\hat{f}_i \hat{\rho}))$, $i = 1, \dots, 4$, where $\hat{\rho}$ is the density matrix of the system, accounting for the longitudinal relaxation accompanied by population transfer between the levels. Defining the matrices $\hat{f}_i$, according to:

$$\begin{aligned}
\hat{\Gamma}_1 &= \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & \Gamma_1 & 0 & 0 \\ 0 & 0 & \gamma & 0 \\ 0 & 0 & 0 & \Gamma_3 \end{pmatrix}; \hat{\Gamma}_2 = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & -(\Gamma_1 + \Gamma_2) & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \gamma \end{pmatrix} \\
\hat{\Gamma}_3 &= \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & \Gamma_2 & 0 & 0 \\ 0 & 0 & -\gamma & 0 \\ 0 & 0 & 0 & \Gamma_1 \end{pmatrix}; \hat{\Gamma}_4 = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -(\Gamma_1 + \Gamma_3 + \gamma) \end{pmatrix}
\end{aligned} \tag{8.9}$$

$\hat{\sigma}$ -matrix components can be expressed in terms of real state vector components as follows:

$$\begin{aligned}
\sigma_{11} &= \frac{1}{12} \left(2\sqrt{3}S_{14}(2\gamma - \Gamma_1) - \sqrt{6}S_{15}(\gamma + \Gamma_1 - 3\Gamma_3) + 3(\gamma + \Gamma_1 + 2S_{13}\Gamma_1 + \Gamma_3) \right) \\
\sigma_{22} &= \frac{1}{12} \left(3\gamma(1 + \sqrt{6}S_{15}) + (-3 - 6S_{13} + 2\sqrt{3}S_{14} + \sqrt{6}S_{15})(\Gamma_1 + \Gamma_2) \right) \\
\sigma_{33} &= \frac{1}{12} \left(\sqrt{6}S_{15}(\gamma + 3\Gamma_1 - \Gamma_2) - 2\sqrt{3}S_{14}(2\gamma + \Gamma_2) + 3(-\gamma + \Gamma_1 + \Gamma_2 + 2S_{13}\Gamma_2) \right) \\
\sigma_{44} &= -\frac{1}{4} \left(1 + \sqrt{6}S_{15} \right) (\gamma + \Gamma_1 + \Gamma_3)
\end{aligned} \tag{8.10}$$

The introduction of a dephasing rates matrix $\hat{\Gamma}_t$ accounting for the dissipation in the system due to polarisation relaxation, according to:

$$\hat{\Gamma}_t = \begin{pmatrix} 0 & \Gamma_\mu & \Gamma_\mu & \Gamma_{\mu-1} \\ \Gamma_\mu & 0 & \Gamma_\mu & \Gamma_{\mu-1} \\ \Gamma_\mu & \Gamma_\mu & 0 & \Gamma_{\mu-1} \\ \Gamma_{\mu-1} & \Gamma_{\mu-1} & \Gamma_{\mu-1} & 0 \end{pmatrix} \tag{8.11}$$

implies transverse relaxation (dephasing) times in the first term of Eq.(8.7) given by: $T_1 = T_2 = T_3 = T_7 = T_8 = T_9 = 1/\Gamma_\mu$, $T_4 = T_5 = T_6 = T_{10} = T_{11} = T_{12} = 1/\Gamma_{\mu-1}$.

Expressions for the longitudinal population relaxation times T_{13}, T_{14}, T_{15} are derived through the second term in Eq.(8.7), giving:

$$T_{13} = \frac{4}{2\Gamma_1 + \Gamma_2}; T_{14} = \frac{4}{2\gamma + \Gamma_2}; T_{15} = \frac{2}{\gamma + \Gamma_1 + \Gamma_3} \tag{8.12}$$

Within the coherent master Maxwell-pseudospin equations approach [34], the one-dimensional Maxwell's curl equations for the circularly polarised laser pulse propagating in an isotropic medium with effective dielectric constant, ϵ , calculated in Sec. 8.2.1:

$$\begin{aligned}
\frac{\partial H_x(z,t)}{\partial t} &= \frac{1}{\mu} \frac{\partial E_y(z,t)}{\partial z} \\
\frac{\partial H_y(z,t)}{\partial t} &= -\frac{1}{\mu} \frac{\partial E_x(z,t)}{\partial z} \\
\frac{\partial E_x(z,t)}{\partial t} &= -\frac{1}{\epsilon} \frac{\partial H_y(z,t)}{\partial z} - \frac{1}{\epsilon} \frac{\partial P_x(z,t)}{\partial t} \\
\frac{\partial E_y(z,t)}{\partial t} &= \frac{1}{\epsilon} \frac{\partial H_x(z,t)}{\partial z} - \frac{1}{\epsilon} \frac{\partial P_y(z,t)}{\partial t}
\end{aligned} \tag{8.13}$$

are solved self-consistently with the pseudospin equations Eq.(8.7). The two sets of equations are coupled through the macroscopic polarisation induced in the medium by the circularly polarised electromagnetic wave. We derive the following relations between the medium polarisation vector and the real state vector components:

$$\begin{aligned} P_x &= -\wp N_a (S_1 + S_6) \\ P_y &= -\wp N_a (-S_7 + S_{12}) \end{aligned} \quad (8.14)$$

where N_a is the density of the ensemble of resonantly absorbing/amplifying four-level systems. These polarizations act as source terms in the vector Maxwell's equation for the optical wave propagation Eq.(8.13).

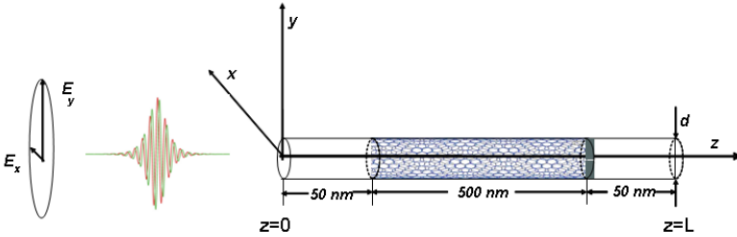


Fig. 8.6 (Color online) Simulation domain: The isolated SWCNT with length $L_n = 500\text{nm}$ is placed between two free space regions, each 50nm -long. The source pulse starts to propagate from the left boundary $z = 0$.

The above set of equations (8.13),(8.7),(8.14), is solved numerically for the fields and the real-vector components in the time domain employing the Finite-Difference Time-Domain (FDTD) technique without invoking any approximations, such as slowly-varying wave approximation (SVEA) and rotating-wave approximation (RWA).

The initial circularly-polarized optical pulsed excitation, applied at the left boundary of the simulation domain $z = 0$ (Fig. 8.6), is modeled by two orthogonal linearly polarized optical waves, phase-shifted by $\pi/2$:

$$\begin{aligned} \sigma^- &\begin{cases} E_x(z=0, t) = E_0 e^{-(t-t_0)^2/t_d^2} \cos(\omega_0 t) \\ E_y(z=0, t) = -E_0 e^{-(t-t_0)^2/t_d^2} \sin(\omega_0 t) \end{cases} \\ \sigma^+ &\begin{cases} E_x(z=0, t) = E_0 e^{-(t-t_0)^2/t_d^2} \cos(\omega_0 t) \\ E_y(z=0, t) = E_0 e^{-(t-t_0)^2/t_d^2} \sin(\omega_0 t) \end{cases} \end{aligned} \quad (8.15)$$

where E_0 is the initial field amplitude, the pulse carrier frequency is tuned in resonance with the dipole optical transition frequency ω_0 and is modulated by a

Gaussian, centered at t_0 with characteristic decay time t_d , which determines the pulse duration. Linearly polarised pulse along x (y) direction is modeled by:

$$\begin{cases} E_x(z=0, t) = E_0 e^{-(t-t_0)^2/t_d^2} \cos(\omega_0 t) \\ E_y(z=0, t) = 0 \end{cases} \quad (8.16)$$

$$\begin{cases} E_x(z=0, t) = 0 \\ E_y(z=0, t) = E_0 e^{-(t-t_0)^2/t_d^2} \sin(\omega_0 t) \end{cases}$$

8.4 Simulation results for the natural optical activity

The simulated semiconducting (5,4) AL-handed SWCNT structure with effective refractive index $n = 2.3$ (see calculation in 8.2.1) is shown in Fig. 8.6 embedded between two free space (air) regions with refractive index $n = 1.00029$. The simulation domain is finely discretized in space with $\Delta z = 1$ nm, which through the Courant numerical stability criterion, corresponds to a time step of $\Delta t = 3.3356 \times 10^{-18}$ s.

The ultrashort optical pulse with pulse duration $\tau = 60$ fs (selected to match the experimental conditions in [33]) and Gaussian envelope is injected from the left boundary ($z=0$). The pulse center frequency $\omega_0 = 3.0066 \times 10^{15}$ rad.s $^{-1}$ is tuned in resonance with the energy splitting between $\mu \rightarrow \mu \pm 1$ subband states of $E_{\mu, \mu \pm 1} \sim 1.9815$ eV, corresponding to a wavelength $\lambda = 626.5$ nm. Since we are interested in the high-intensity nonlinear regime, throughout the simulations the pulse area below the pulse envelope is chosen to be π , giving initial electric field amplitude $E_0 = 6.0977 \times 10^8$ Vm $^{-1}$ (see Eq.8.15). A π -pulse is of particular interest since it completely inverts the population in a two-level system. In order to ensure that our ensemble approach describes properly the single nano-object, we select the density of the coupled oscillators, described by four-level systems, in such a way that the simulated nanotube volume (with diameter $d = 0.61145$ nm and length $L_n = 500$ nm) contains on average only one carbon nanotube [34]. This yields an average density $N_a = 6.811 \times 10^{24}$ m $^{-3}$ of resonant absorbers.

The ultrafast optically-induced polarisation dynamics depends very strongly on the phenomenological relaxation rates. Assuming the value $\wp = 3.613 \times 10^{-29}$ Cm calculated in Sec. 8.2.2 for the optical dipole matrix element, we use the expression for the spontaneous emission rate [41], taking into account the corresponding energy separation for each transition, to obtain an estimate for the relaxation rates. Thus we obtain the following parameters: $\Gamma_1 = 2.90729$ ns $^{-1}$, $\Gamma_2 = 9.81211$ ns $^{-1}$, $\Gamma_3 = 1.22651$ ns $^{-1}$. We take the experimental value obtained in [33] for the intra-band optical transitions, namely $\gamma = 130$ fs $^{-1}$. Since the dephasing rates for the states involved are largely unknown, we treat them as phenomenological parameters, adopting the following values: $\Gamma_\mu = 800$ fs $^{-1}$ and $\Gamma_{\mu-1} = 1.6$ ps $^{-1}$. Throughout the simulations the population density of all four levels is conserved ($\rho_{11} + \rho_{22} +$

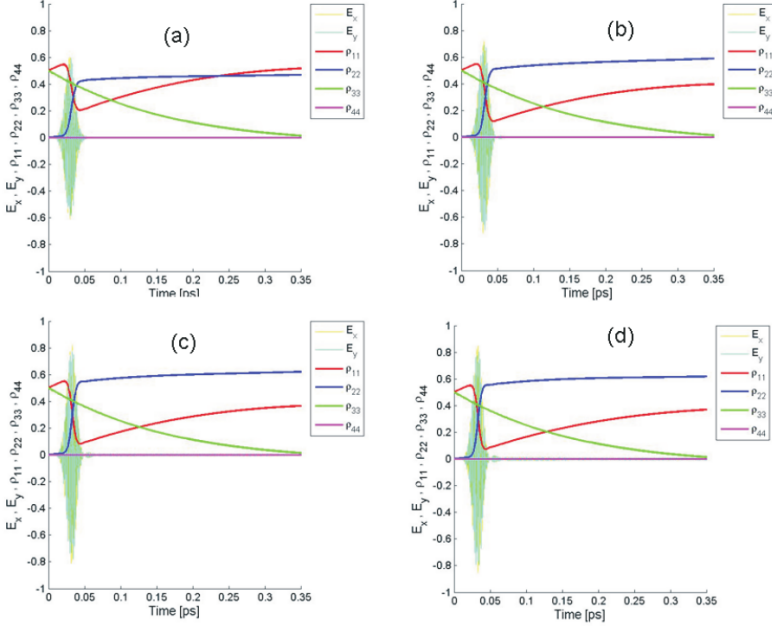


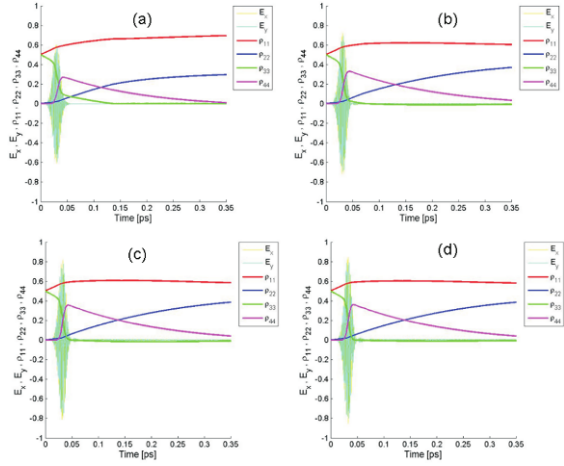
Fig. 8.7 Time evolution of the electric field vector components E_x, E_y and the populations of all four levels in Fig. 8.5 under right circularly polarised optical pulse excitation σ^+ at four different locations along the nanotube axis (a) $z = 175$ nm; (b) $z = 300$ nm; (c) $z = 425$ nm; (d) $z = 550$ nm; measured from the left boundary $z = 0$ in Fig. 8.6. The initial population is assumed equally distributed between the lower-lying levels $|1\rangle$ and $|3\rangle$: $\rho_{11i} = \rho_{33i} = 1/2$. Level $|4\rangle$ is not involved in the dynamics.

$\rho_{33} + \rho_{44} = 1$). We consider separately the two cases of an ultrafast resonant optical excitation of the $|1\rangle \rightarrow |2\rangle$ transition with σ^+ helicity and of the $|3\rangle \rightarrow |4\rangle$ transition with σ^- helicity (Fig. 8.5). Note that in the former case level $|4\rangle$ does not participate in the relaxation dynamics and the system is effectively a three-level Λ -system, rather than a four-level system. The ultrashort circularly polarised source pulse is injected into the medium and the temporal dynamics of the electric field vector components and population of all four levels are sampled at four different locations along the nanotube z -axis. The time evolution of the pulse E-field components amplitude, normalised with respect to E_0 , or the pulse reshaping and the level populations are shown in Fig. 8.7 for times immediately after the excitation. The initial population is assumed equally distributed between the lower-lying levels ($\rho_{11i} = \rho_{33i} = 1/2$). The population of the lowest level $|1\rangle$, initially slightly increases due to the ultrafast population transfer from level $|3\rangle$, which is initially populated. The optical pulse excites the population to level $|2\rangle$: the population of level $|1\rangle$ sharply decreases while at the same time the population of level $|2\rangle$ increases. The population of level $|3\rangle$ decays almost exponentially, since the pumping of this level through population relaxation from $|2\rangle$ is at a much slower rate (by three orders of

magnitude) than the intraband relaxation rate. The pulse trailing edge is slightly distorted due to the dipole formed between levels $|3\rangle$ and $|2\rangle$ (at the intersection point between ρ_{22} and ρ_{33} , Fig. 8.7), re-radiating back to the field. It is obvious that the final population after very long simulation times will relax to the equilibrium value $\rho_{11e} = 1$ into level $|1\rangle$ (not shown on this time scale). During the pulse propagation along the nanotube, the pulse is undergoing resonant absorption and emission resulting in pulse amplitude amplification, increasing from ~ 0.6 near the left boundary to ~ 0.8 at the right structure boundary (Fig. 8.7(a),(d)). This implies that the carbon nanotube behaves as a laser gain medium under an ultrashort circularly polarised pulsed optical excitation.

The system dynamics under σ^- polarised optical excitation is quite distinct, since

Fig. 8.8 Time evolution of the electric field vector components E_x, E_y and the populations of all four levels in Fig. 8.5 under left circularly polarised optical pulse excitation σ^- at four different locations along the nanotube axis (a) $z = 175$ nm; (b) $z = 300$ nm; (c) $z = 425$ nm; (d) $z = 550$ nm; measured from the left boundary $z = 0$ in Fig. 8.6.



all four levels are involved in it (Fig. 8.8). As in the previous case, the initial population is assumed equally distributed between the lower-lying levels. The population residing in level $|3\rangle$ is partly resonantly excited into level $|4\rangle$ by the passage of the pulse, shown by the sharp increase of ρ_{44} and a corresponding decrease of ρ_{33} . The population of level $|1\rangle$ slowly increases, rather than sharply dropping (cf. Fig. 8.7), due to population transfer by spontaneous emission, interband ($\mu - 1 \rightarrow \mu - 1$) and intraband relaxation processes (Fig. 8.5). The population of level $|2\rangle$ increases at a slower rate during the passage of the pulse, due to depletion by the competing relaxation channels, reaching a maximum at later times (not shown) and subsequently decreasing. Eventually the whole population relaxes into the lowest-lying state $|1\rangle$. As in the previous case considered, a slight pulse trailing edge distortion is observed, due to forming of a dipole between levels $|4\rangle$ and $|2\rangle$, and $|3\rangle$ and $|2\rangle$. Similar to the previous case, the pulse amplitude is amplified during the pulse propagation along the nanotube axis. It is obvious that the different dynamics involved in the two cases of ultrafast optical excitation by σ^+ and σ^- polarised pulse, would lead to an asymmetry of the optical properties and therefore to a circular dichroism, birefrin-

gence and optical rotation. In what follows we shall evaluate the magnitude of these chirality effects.

Fig. 8.9 (a) Time trace of the E_x component of the input (solid blue line) and output (solid red line) σ^+ polarised pulse; (b) Transmission (Fourier) spectra vs wavelength of the input (blue line) and output (red line) σ^+ pulse; (c) Time trace of the E_y component of the input (blue line) and output (red line) σ^+ polarised pulse; (d) Transmission spectra vs wavelength of the input (blue line) and output (red line) pulses.

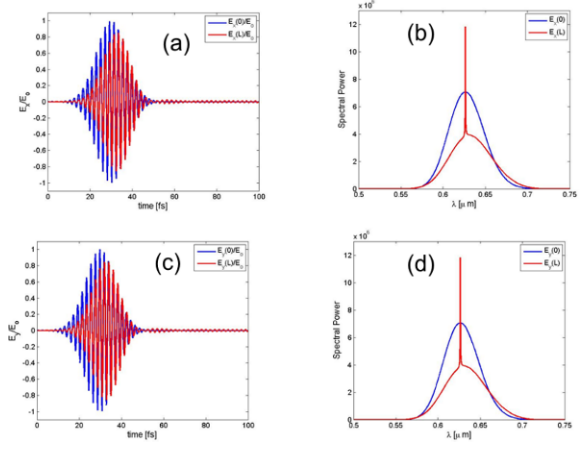
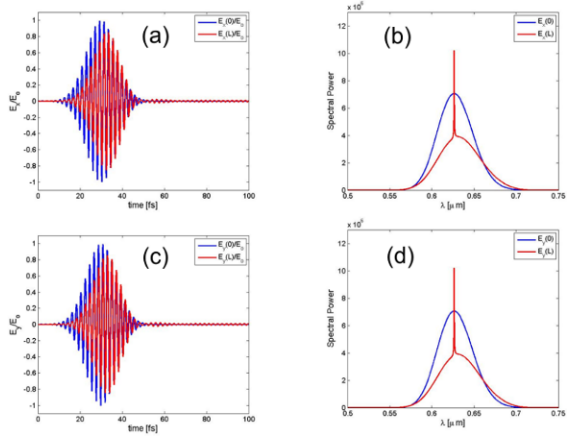


Fig. 8.10 (a) Time trace of the E_x component of the input (solid blue line) and output (solid red line) σ^- polarised pulse; (b) Transmission (Fourier) spectra vs wavelength of the input (blue line) and output (red line) σ^- pulse; (c) Time trace of the E_y component of the input (blue line) and output (red line) σ^- polarised pulse; (d) Transmission spectra vs wavelength of the input (blue line) and output (red line) pulses.



From experimental point of view, it would be of particular interest to be able to distinguish between the spectra of the transmitted pulses at the output facet of the simulated domain for each helicity of the ultrashort excitation pulse. The time traces of the E_x and E_y electric field components of the input and transmitted pulse (at the output facet of the simulated structure, Fig. 8.6) and their respective Fourier spectra are shown in Fig. 8.9 for the case of an ultrashort σ^+ polarised excitation. The transmission spectra exhibit a sharp peak at the resonant wavelength, which is an indication of resonant amplification. The corresponding time traces and Fourier spectra for σ^- polarised excitation are shown in Fig. 8.10. Similar to the previous case, the

transmission spectrum exhibits a sharp resonant peak at the resonant wavelength, however a comparison between Fig. 8.9 and 8.10 reveals much higher intensity of the peak, corresponding to the σ^+ excitation. The difference in the ultrafast transient response can be exploited in an experiment aiming to unambiguously determine the chirality of a single carbon nanotube by using ultrafast circularly polarised pulses of both helicities. In what follows, we shall demonstrate that the difference in the polarisation-resolved (along x and y) transmission spectra of a linearly-polarised excitation is much more pronounced and permits identifying the precise helical for of the SWCNT.

In order to numerically demonstrate the rotation of the polarisation plane during the pulse propagation across the medium, a linearly polarised pulse (Eq.8.16, first line) is injected. The system temporal dynamics induced by the ultrashort linearly polarised pulse is shown in Fig. 8.11.

Fig. 8.11 Time evolution of the electric field vector components E_x, E_y and the populations of all four levels in Fig. 8.5 under linearly polarised optical pulse excitation at four different locations along the nanotube axis (a) $z = 175$ nm; (b) $z = 300$ nm; (c) $z = 425$ nm; (d) $z = 550$ nm; measured from the left boundary $z = 0$ in Fig. 8.6. The initial population is assumed equally distributed between the lower-lying levels $|1\rangle$ and $|3\rangle$: $\rho_{11i} = \rho_{33i} = 1/2$.

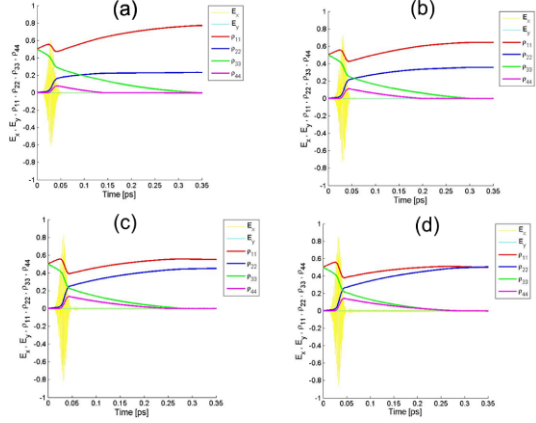
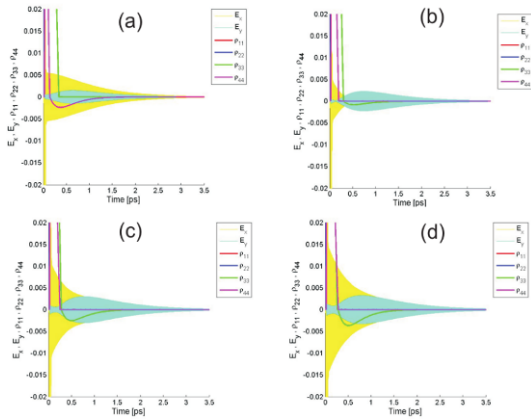


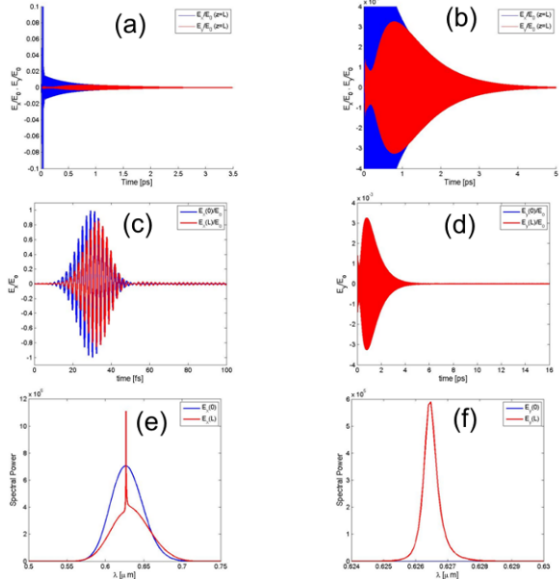
Fig. 8.12 Expanded view of Fig. 8.11 showing the time evolution of the electric field vector components E_x, E_y and the populations of all four levels under linearly polarised optical pulse excitation at four different locations along the nanotube axis (a) $z = 175$ nm; (b) $z = 300$ nm; (c) $z = 425$ nm; (d) $z = 550$ nm; measured from the left boundary $z = 0$ in Fig. 8.6.



Note that on this scale amnonvanishing E_y component is not apparent, however on an expanded scale (Fig. 8.12) the appearance of a second E_y component (cyan line), and therefore the optical rotation of the E-field vector polarisation is clearly visible. The amplitude of E_y component continuously increases as the pulse propagates along the nanotube structure.

We should therefore expect a maximum optical rotation angle at the output facet, shown in Fig. 8.13 (a,b). The transmitted pulse is elliptically polarised. The optical transmission spectra of the x-polarised and y-polarised electric field vector components are quite distinct: while the former exhibits a sharp resonant peak superimposed on a broader line, the latter exhibits a single spectral line centered at the resonant wavelength.

Fig. 8.13 (a) Time evolution of the E_x (blue line) and E_y (red line) components at the output facet of the simulated domain (E_y is initially set to 0 to model linearly polarised pulse); (b) Expanded view of (a) clearly showing the build-up with time of the electric field vector E_y component amplitude at the output facet (c) Time trace of the E_x component at the input facet ($z = 0$) and at the output facet ($z = L$); (d) Time trace of the E_y component at the input ($E_y = 0$) and at the output ($z = L$) facet; (e) Fourier (transmission) spectra of the time traces in (c); (f) Fourier spectrum of $E_y(z = L)$ at the output facet.



In order to obtain a quantitative estimate of the natural optical activity in a single chiral carbon nanotube, we calculate the average absorption/gain coefficient and phase shift induced by the resonant medium. The former allows us to calculate the magnitude of the circular dichroism and the latter represents a measure of the rotation angle. Consider two locations z_1 and z_2 separated by one dielectric wavelength $l = z_2 - z_1 = \lambda_0/n$, where λ_0 is the resonant wavelength. The complex propagation factor of a resonant optical pulse propagating in an amplifying/absorbing medium from z_1 to z_2 is given by:

$$\begin{aligned} e^{ik_c(z_2-z_1)} &= \frac{E_x(z_2, \omega)}{E_x(z_1, \omega)} = z_x \\ e^{ik_c(z_2-z_1)} &= \frac{E_y(z_2, \omega)}{E_y(z_1, \omega)} = z_y \end{aligned} \quad (8.17)$$

for the E_x and E_y electric field components, respectively. The wavevector $k_c = \beta + i\gamma$, where β is the phase shift per unit length in the optical pulse induced by the interaction with the resonant medium and γ is the absorption/gain coefficient. The following expressions for β and γ can be easily obtained:

$$\begin{aligned}\gamma_{x,y} &= -\frac{\ln|z_{x,y}|}{l} \\ \beta_{x,y} &= \frac{1}{l} \arctan\left(\frac{\text{Im}(z_{x,y})}{\text{Re}(z_{x,y})}\right)\end{aligned}\quad (8.18)$$

We select four pairs of points z along the nanotube length with staggered distances from the left boundary ($z = 0$) within the carbon nanotube structure, separated by one dielectric wavelength and calculate the above quantities for each pair.

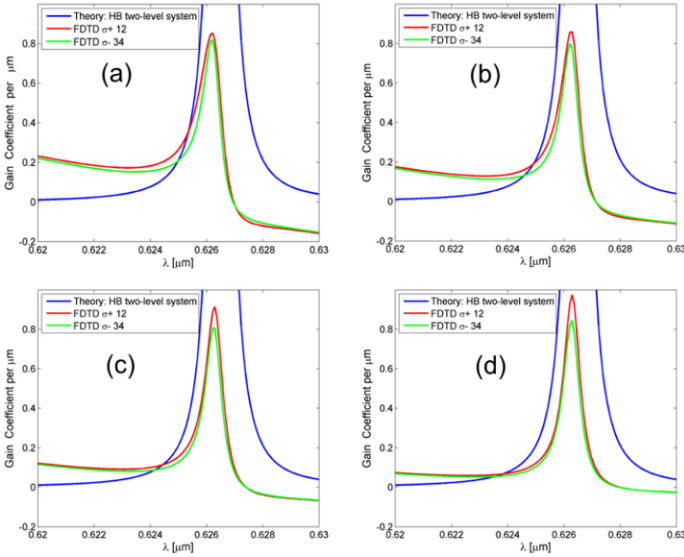


Fig. 8.14 Spatially resolved calculated gain coefficient per micron vs wavelength for $E_x(E_y)$ electric field vector component of a σ^+ (red line) and σ^- (green line) circularly polarised ultrashort optical excitation and the theoretical gain coefficient of a homogeneously broadened resonant two-level system (blue line) for (a) pair of points z_1 and z_2 within the carbon nanotube separated by one dielectric resonant wavelength. The offset of the first point z_1 is 5 nm from the beginning of the nanotube structure modeled as a absorption/gain medium: $\Delta A = 0.033785 \mu\text{m}^{-1}$; (b) a pair of points z_3 and z_4 within the carbon nanotube, one dielectric length apart, z_3 is shifted to the right by 1 nm with respect to z_1 : $\Delta A = 0.060104 \mu\text{m}^{-1}$; (c) a pair of points z_5 and z_6 within the active medium, separated by one dielectric wavelength, z_5 is shifted by 1 nm to the right of z_3 : $\Delta A = 0.107539 \mu\text{m}^{-1}$; (d) a pair of points z_7 and z_8 within the active medium, separated by one dielectric wavelength, z_7 is shifted by 1 nm to the right of z_5 : $\Delta A = 0.1291 \mu\text{m}^{-1}$;

The plots in Fig. 8.14 show a comparison between the gain coefficients for σ^+ excitation, corresponding to pumping the $|1\rangle \rightarrow |2\rangle$ transition and for σ^- excitation, corresponding to pumping the $|3\rangle \rightarrow |4\rangle$ transition in Fig. 8.5. The theoretical gain

coefficient of a homogeneously broadened two-level system is shown on the same plot for reference. The difference between the two plots at the resonant wavelength gives us an estimate of the magnitude of the natural circular dichroism in the (5,4) carbon nanotube. The spatially resolved circular dichroism along the nanotube axis is calculated from the difference between the maxima of the gain coefficients at the resonant wavelength, corresponding to excitation with the two helicities (R or σ^+) and (L or σ^-): $\Delta A = G_R - G_L$ at each location (see Fig. 8.14), giving an average of $0.083 \mu\text{m}^{-1}$. This result is in good agreement with the theoretically predicted natural circular dichroism (cf. [35] Eq. (44)), giving a value of $1.03 \mu\text{m}^{-1}$ for the specific (5,4) nanotube considered. By comparison, the absolute value of the circular dichroism of an artificial helicoidal bilayered structure [28] varies in the range 5 – 9 dB which is equivalent to a linear absorption/gain coefficient in the range $1.15 - 2.07 \mu\text{m}^{-1}$.

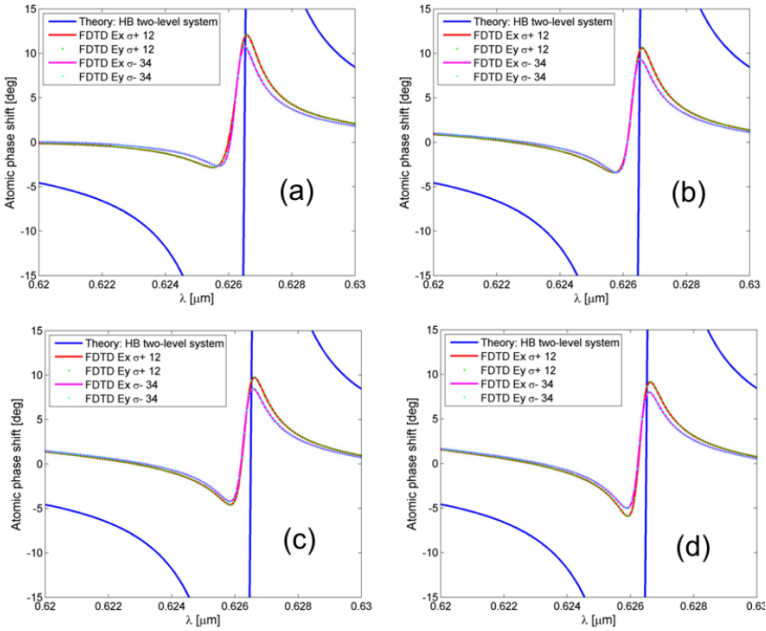
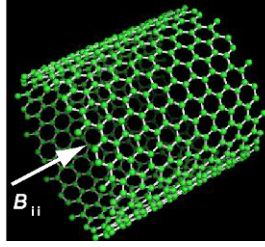


Fig. 8.15 Spatially resolved calculated phase shift vs wavelength for E_x (red line) and E_y (green dot) components under σ^+ excitation and for E_x (magenta line) and E_y (cyan dot) under σ^- circularly polarised ultrashort optical excitation and the theoretical phase shift of a homogeneously broadened resonant two-level system (blue line) for the cases in Fig. 8.14: (a) $\Delta\phi = \beta l = 1.457076^\circ$, $\rho = 2.6746^\circ/\mu\text{m}$; (b) $\Delta\phi = 1.3388^\circ$, $\rho = 2.4574^\circ/\mu\text{m}$; (c) $\Delta\phi = 1.64222^\circ$, $\rho = 3.0145^\circ/\mu\text{m}$; (d) $\Delta\phi = 2.016985^\circ$, $\rho = 3.7024^\circ/\mu\text{m}$;

The phase shift difference $\Delta\phi = \Delta\phi_R - \Delta\phi_L$ introduced by the nonsymmetric system response under σ^+ (transition $|1\rangle \rightarrow |2\rangle$) and σ^- (transition $|3\rangle \rightarrow |4\rangle$) circularly polarised pulse excitation represents a measure for the rotation angle. The result of the

Fig. 8.16 Scheme of a single chiral nanotube (20, 10) threaded by an axial magnetic field



polarisation is to change the phase delay per unit length from $\beta_{R(L)}$ to $\beta_{R(L)} + \Delta\beta_{R(L)}$. The atomic phase shift ($\beta_{R(L),x(y)}l$) for the four cases described above, is plotted in Fig. 8.15 and the rotation angle and the specific rotatory power per unit length: $\rho = \pi(n_L - n_R)/\lambda_0$, where n_L and n_R are the refractive indices for left- and right-circularly polarised light, respectively, are given for each case. The specific rotatory power varies in the interval from $\rho = 2.46 - 3.7^\circ/\mu\text{m}$, giving an average value of $\sim 2962.24^\circ/\text{mm}$. The corresponding anisotropy of the refractive indices for left- and right-circularly polarised light is on the order of $\Delta n = n_L - n_R = 0.0103$. The calculated natural polarisation rotatory power, for the special case of a (5, 4) nanotube considered, is exceeding the giant gyrotropy reported in artificial photonic metamaterials of up to $2500^\circ/\text{mm}$ [27]. We should note, however, that the complexity of the carbon nanotube molecular structure allows for engineering the optical activity in a wide range. By comparison, the optical activity of e.g. quartz illuminated by the D line of sodium light ($\lambda = 589.3 \text{ nm}$), is $21.7^\circ/\text{mm}$, implying refractive indices difference $|n_L - n_R| \sim 7.1 \times 10^{-5}$; $\rho = 32.5^\circ/\text{mm}$ for cinnabar (HgS). A comparison of the specific rotatory power for a group of crystals is given in Ref.[50] (Table II) showing a wide range of variation from $2.24^\circ/\text{mm}$ for NaBrO_3 to $522^\circ/\text{mm}$ for AgGaS_2 . Liquid substances exhibit much lower values of specific rotatory power, e.g. $\rho = -0.37^\circ/\text{mm}$ for turpentine ($T = 10^\circ$, $\lambda = 589.3 \text{ nm}$); $\rho = 1.18^\circ/\text{mm}$ for corn syrup, etc. Cholesteric liquid crystals and sculptured thin films exhibit large rotatory power in the visible spectrum $\sim 1000^\circ/\text{mm}$ [43], and $\sim 6000^\circ/\text{mm}$ [44], respectively.

8.5 Magneto-optical activity of a chiral SWCNT in an axial magnetic field

We shall now focus on the theoretical description of the resonant coherent nonlinear optical activity when a static magnetic field $B_{\parallel}$ is threading the nanotube (Fig. 8.16). The rotation of the polarisation of a plane-polarised electromagnetic wave propagating in a substance under a static magnetic field along the direction of propagation is known as Faraday rotation. In the presence of an axial magnetic field the electronic band structure of a single carbon nanotube, and the electronic states near the band gap edge in particular, significantly change, owing to the combined action of

two effects: the spin-B interaction resulting in Zeeman splitting of the energy levels [13, 46, 15] and the appearance of the Aharonov-Bohm phase in the wave function [31, 14, 45, 47, 16] resulting in an additional energy level shift. The two symmetric subbands at K (K') point of the Brillouin zone are degenerate at $B_{\parallel} = 0$. An applied magnetic field along the nanotube axis lifts this degeneracy and shifts the energy levels. As a result the energy gap of one of the subbands (K') becomes larger while the energy gap of the other subband (K) becomes smaller [46]. Furthermore, the possibility of a magnetic field-induced metal-insulator transition has been put forward [49]. It has been theoretically predicted that the effect of the Aharonov-Bohm (AB) flux on the energy gap is to induce oscillations between zero and a fixed value with a period of the flux quantum $\Phi_0 = h/e$ [31, 15, 48] resulting in periodical oscillations of the magneto-optical absorption spectra. At a fixed value of the static magnetic field, however, the orbital AB effect leads to a uniform shift in the energy levels.

8.5.1 Theoretical model of the nonlinear Faraday rotation in an axial magnetic field

Without loss of generality, we shall consider the electronic states near the band gap at the K-point in the Brillouin zone. Therefore the overall effect of the two mechanisms described above will be a band gap reduction. We consider and calculate separately the contributions to the band gap in respect of the Zeeman splitting and the Aharonov-Bohm effect. The Zeeman splitting, or the spin-B interaction energy is given by:

$$E_z = \mu_B g_e \sigma B_{\parallel} \quad (8.19)$$

where $\mu_B = \frac{e\hbar}{2m_e}$ is the Bohr magneton, the electron g-factor, g_e is taken to be the same as that of pure graphite (~ 2), $\sigma = \pm 1/2$ is the z-axis projection of the electron spin (spin up/spin down state), and m_e is the free electron mass. For a magnetic field $B_{\parallel} = 8T$, the above equation gives an energy shift of $E_z \approx 0.46 meV$, which corresponds to a resonant frequency $\omega_z = 7.026 \times 10^{11} rad/s$. The predicted oscillatory magnetic field dependence of the energy gap of a semiconducting nanotube ([32, 13]) is of the form:

$$E_G(\Phi) = \begin{cases} 3E_G(0) \left| \frac{1}{3} - \frac{\Phi}{\Phi_0} \right|, & 0 \leq \Phi/\Phi_0 \leq 1/2, \\ 3E_G(0) \left| \frac{2}{3} - \frac{\Phi}{\Phi_0} \right|, & 1/2 \leq \Phi/\Phi_0 \leq 1 \end{cases} \quad (8.20)$$

where $E_G(0) = \hbar\omega_0$ is the energy gap at zero magnetic field ($B = 0$), ω_0 is the resonant transition frequency. For the case of a magnetic field $B = 8T$ with a flux Φ threading the (5,4) nanotube cross-section with a diameter $d = 0.61145$ nm, the ratio $\Phi/\Phi_0 = 0.00057$, and therefore the first of the equations above holds. This leads to an energy level shift, or band gap renormalisation due to the orbital AB effect of $E_{AB} = 3.37$ meV, corresponding to a resonant angular frequency $\omega_{AB} =$

5.11655×10^{12} rad/s. In an external magnetic field the energy levels near the band gap of Fig. 8.5 split and the spin degeneracy is lifted (see Fig. 8.17).

The energy level system can be split into two reduced system of levels, each of

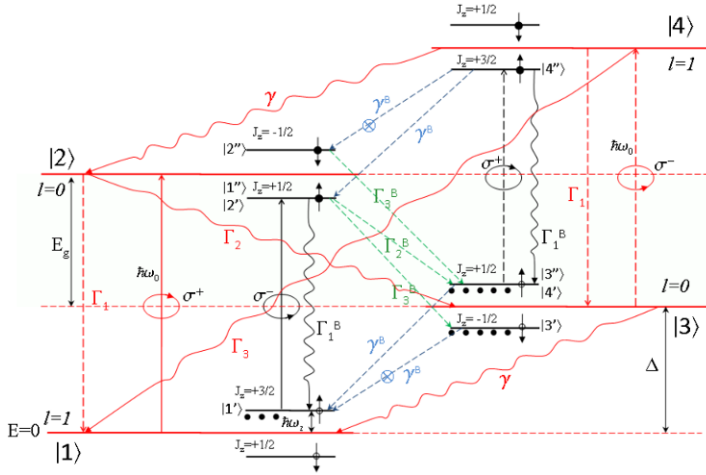


Fig. 8.17 Zeeman splitting ($\hbar\omega_z$) of the energy levels near the band gap of a (5,4) nanotube in an external axial magnetic field. The original set of levels ($|1\rangle - |4\rangle$) is labeled by the (quasi-angular) orbital momentum quantum number $l = 0, 1$; the resulting energy levels are labeled by the projection of the total angular momentum $J = l + s$ along the nanotube axis (z). Two reduced sets of levels, that energetically are nearest to the band edge, can be identified, namely $|1'\rangle - |4'\rangle$ and $|1''\rangle - |4''\rangle$ (note that level $|4'\rangle$ and $|3''\rangle$ are common for the two systems). The energy band gap E_g is shaded, Δ is the energy separation between the $l = 0$ and $l = 1$ states (assumed, for simplicity, equal for the valence and conduction band states). The original system of levels, the stimulated optically pumped transitions by $\sigma^{+(-)}$ light and relaxation processes are plotted in red. The black solid (dashed) arrows denote the resonant excitation of $|1'\rangle \rightarrow |2'\rangle$ ($|3''\rangle \rightarrow |4''\rangle$) transition by σ^- (σ^+) light. Note that the initial population (small black circles) is equally distributed between levels $|1'\rangle$ and $|3'\rangle$ in the valence band for the first ($'$) reduced system of levels, whereas the total population is residing in level $|3''\rangle$ in the valence band of the second ($''$) reduced system of levels. The spontaneous emission rates, designated by wavy lines, and the relaxation rates, are modified in a magnetic field, which is reflected by the superscript B . The small black arrows denote the spin-up and spin-down states; note that the spin-up state is energetically higher than the spin-down state in the conduction band, whereas the opposite is valid for the valence band states. The forbidden transitions by the dipole optical selection rules are designated by $\otimes$.

which represents a mirror image of the other, however the symmetry is broken by the allowed optical transitions in each case. For simplicity, the Aharonov-Bohm uniform shift ($\hbar\omega_{AB}$) is not shown in Fig. 8.17. Analogously to the $B = 0$ case, the system Hamiltonian for an AL-handed SWCNT in an axial magnetic field $B \neq 0$ (for the ($'$)-system), excited by a σ^- pulse can be written in the form;

$$\hat{H}' = \hbar \begin{pmatrix} \omega_z & -\frac{1}{2}(\Omega_x - i\Omega_y) & 0 & 0 \\ -\frac{1}{2}(\Omega_x + i\Omega_y) & \omega_0 - \omega_z & 0 & 0 \\ 0 & 0 & \Delta - \omega_z & 0 \\ 0 & 0 & 0 & \Delta + \omega_z \end{pmatrix} \quad (8.21)$$

where we have made the band gap renormalisation with the AB shift: $\omega_0 \rightarrow \omega_0 - \omega_{AB}$, giving a torque vector:

$$\gamma = \left(-\Omega_x, 0, 0, 0, 0, 0, -\Omega_y, 0, 0, 0, 0, 0, \omega_0 - 2\omega_z, \frac{2\Delta - \omega_0 - 2\omega_z}{\sqrt{3}}, \frac{2\Delta - \omega_0 + 4\omega_z}{\sqrt{6}} \right) \quad (8.22)$$

The time evolution of the four-level quantum system under an external time-dependent dipole coupling perturbation is given by Eq. 8.7. Similar to Sec. 8.2, $\hat{I}_i$ matrices are introduced, according to:

$$\begin{aligned} \hat{I}_1 &= \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & I_1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \gamma \end{pmatrix}; \hat{I}_2 = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & -(I_1 + I_2 + I_3) & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \\ \hat{I}_3 &= \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & I_3 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}; \hat{I}_4 = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & I_2 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -\gamma \end{pmatrix} \end{aligned} \quad (8.23)$$

resulting in the following expressions for the diagonal components of the matrix $\hat{\sigma}$, expressed in terms of the real state vector components:

$$\begin{aligned} \sigma_{11} &= \frac{1}{12} \left(3\gamma + \sqrt{6}S_{15}(3\gamma - I_1) + (3 + 6S_{13} - 2\sqrt{3}S_{14})I_1 \right) \\ \sigma_{22} &= -\frac{1}{12} \left(3 + 6S_{13} - 2\sqrt{3}S_{14} - \sqrt{6}S_{15} \right) (I_1 + I_2 + I_3) \\ \sigma_{33} &= \frac{1}{12} \left(3 + 6S_{13} - 2\sqrt{3}S_{14} - \sqrt{6}S_{15} \right) I_3 \\ \sigma_{44} &= \frac{1}{12} \left(-3\gamma + (3 + 6S_{13} - 2\sqrt{3}S_{14})I_2 - \sqrt{6}S_{15}(3\gamma + I_2) \right) \end{aligned} \quad (8.24)$$

The dephasing rate matrix $\hat{I}_i$ is a traceless matrix with all off-diagonal components equal to I_μ , i.e. the transverse relaxation times $T_1 = T_2 = \dots = T_{12} = 1/I_\mu$. The longitudinal relaxation times appearing in the second of the Eq.8.7 are given by:

$$T_{13} = \frac{4}{2I_1 + I_2 + I_3}; T_{14} = \frac{12}{I_2 + 3I_3}; T_{15} = \frac{6}{3\gamma + I_2} \quad (8.25)$$

The macroscopic medium polarisation vector components for this case are given by:

$$\begin{aligned} P_x &= -\beta N_a S_1 \\ P_y &= -\beta N_a S_7 \end{aligned} \quad (8.26)$$

and the resulting pseudospin equations are solved self-consistently with vector Maxwell equations Eq. 8.13 directly in the time domain. The second (") reduced-level system (Fig. 8.17) is described by the following Hamiltonian:

$$\hat{H}'' = \hbar \begin{pmatrix} \omega_0 - \omega_z & 0 & 0 & 0 \\ 0 & \omega_0 + \omega_z & 0 & 0 \\ 0 & 0 & \Delta + \omega_z & -\frac{1}{2}(\Omega_x + i\Omega_y) \\ 0 & 0 & -\frac{1}{2}(\Omega_x - i\Omega_y) & \Delta + \omega_0 - \omega_z \end{pmatrix} \quad (8.27)$$

resulting in the torque vector of the form:

$$\gamma = \left(0, 0, 0, 0, 0, -\Omega_x, 0, 0, 0, 0, 0, \Omega_y, 2\omega_z, \frac{2(\Delta - \omega_0 + \omega_z)}{\sqrt{3}}, \frac{2\Delta + \omega_0 - 4\omega_z}{\sqrt{6}} \right) \quad (8.28)$$

The corresponding diagonal longitudinal relaxation rates matrix $\hat{\sigma}$ is given by:

$$\begin{aligned} \sigma_{11} &= \frac{1}{12} \left(3\gamma + (-3 + 6S_{13} + 2\sqrt{3}S_{14}) I_2 + \sqrt{6}S_{15} (3\gamma + I_2) \right) \\ \sigma_{22} &= -\frac{1}{12} \left(3 + 6S_{13} - 2\sqrt{3}S_{14} - \sqrt{6}S_{15} \right) I_3 \\ \sigma_{33} &= \frac{1}{12} \left(3I_1 + \sqrt{6}S_{15} (3I_1 - I_2) + (3 - 6S_{13} - 2\sqrt{3}S_{14}) I_2 \right) \\ \sigma_{44} &= -\frac{1}{4} \left(1 + \sqrt{6}S_{15} \right) (\gamma + I_1) \end{aligned} \quad (8.29)$$

obtained with the introduction of the following $\hat{I}_i$ matrices:

$$\begin{aligned} \hat{I}_1 &= \begin{pmatrix} -I_2 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \gamma \end{pmatrix}; \hat{I}_2 = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & -I_3 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \\ \hat{I}_3 &= \begin{pmatrix} I_2 & 0 & 0 & 0 \\ 0 & I_3 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & I_1 \end{pmatrix}; \hat{I}_4 = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -(\gamma + I_1) \end{pmatrix} \end{aligned} \quad (8.30)$$

The transverse relaxation rates matrix is given by Eq.8.11, giving $T_1 = T_2 = T_3 = T_7 = T_8 = T_9 = 1/\Gamma_\mu$, and $T_4 = T_5 = T_6 = T_{10} = T_{11} = T_{12} = 1/\Gamma_{\mu-1}$. The following expressions for the longitudinal relaxation rates are obtained:

$$T_{13} = T_{14} = \frac{4}{I_2 + I_3}; T_{15} = \frac{2}{\gamma + I_1} \quad (8.31)$$

The macroscopic polarisation vector components are given by:

$$\begin{aligned} P_x &= -\wp N_a S_6 \\ P_y &= -\wp N_a S_{12} \end{aligned} \quad (8.32)$$

8.5.2 Simulation results for Faraday rotation

The simulated structure is the same as the one described in Fig. 8.6. We resonantly excite $|1'\rangle \rightarrow |2'\rangle$ ($|3''\rangle \rightarrow |4''\rangle$) transition by left (right) circularly polarised σ^- (σ^+) π -pulse with pulse duration $\tau = 60$ fs. The pulse central frequency is the resonant transition frequency, $\omega_0 - \omega_{AB} - 2\omega_z$ and the pulse envelope is a Gaussian function. Owing to the combined effect of the Aharonov-Bohm and Zeeman energy levels shift, the dipole matrix element is modified. An estimate of the optical dipole matrix element in an axial magnetic field can be obtained from the theory developed in [35], taking into account the band gap reduction at $B = 8$ T, giving $\wp = 3.62054 \times 10^{-29}$ Cm. Note that this value is slightly different from the zero magnetic field value (see Sec. 8.4). We recalculate the relaxation times using the above magnetic field dipole coupling constant, thus obtaining the following relaxation rates: $\Gamma_1 = 2.91362 \text{ ns}^{-1}$, $\Gamma_2 = 9.79017 \text{ ns}^{-1}$, $\Gamma_3 = 9.76959 \text{ ns}^{-1}$. The intraband relaxation rate $\gamma = 130 \text{ fs}^{-1}$, and the dephasing rates $\Gamma_\mu = 800 \text{ fs}^{-1}$, $\Gamma_{\mu-1} = 1.6 \text{ ps}^{-1}$ are taken the same as for the zero field case. Note that the initial boundary conditions are different for the excitation by σ^- and σ^+ pulse. While in the former case the initial population is assumed equally distributed between the valence band levels ($\rho_{1'1'i} = \rho_{3'3'i} = 1/2$), nearest to the band edge, in the latter case the whole population is in the single valence band state ($\rho_{4'4'i} = 1$). Similar to Sec. 8.4, the time evolution of the electric field vector components and the population of all four levels is sampled at four points along the nanotube and the real and imaginary part of the complex propagation factor Eq. 8.17, giving the phase shift and the absorption/gain coefficient are calculated. The calculated absorption/gain coefficients for resonant excitation by σ^- (σ^+) pulse are plotted on the same graph (Fig. 8.18) and the theoretical gain coefficient of a homogeneously broadened two-level system is plotted for reference. Note that the resonance is shifted towards longer wavelengths due to the band gap reduction. We should point out that the magnetic circular dichroism spectra at $B = 8$ T shown in Fig. 8.18 are quite distinctive from the zero-magnetic field ones (Fig. 8.14) for the natural optical activity. While the σ^- polarised pulse is amplified during the pulse propagation across the nanotube, the σ^+ polarised pulse is absorbed, resulting in a much larger net circular dichroism. The average value of the magnetic circular dichroism is $0.706 \mu\text{m}^{-1}$, an order of magnitude larger than the natural circular dichroism (cf. Sec. 8.4). The different behavior of the calculated gain and absorption spectra under σ^- (σ^+) polarised optical pulse excitation is a direct consequence of the discrete energy level configuration describing the two cases. While the energy-level system for a σ^- excitation is a four-level system, the one corresponding to a σ^+ excitation is a three-level Λ -system, due to level $|2''\rangle$ being completely disconnected from the rest of the levels, owing to the dipole optical selection rules forbidding transitions

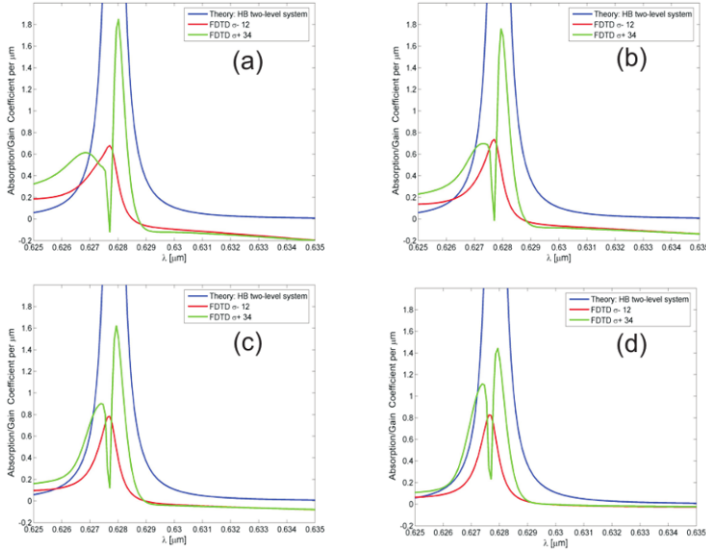


Fig. 8.18 Spatially resolved calculated gain coefficient per micron vs wavelength for $E_x(E_y)$ electric field vector component of a σ^+ (red line) and σ^- (green line) circularly polarised ultrashort optical excitation and the theoretical gain coefficient of a homogeneously broadened resonant two-level system (blue line) at $B = 8$ T for a pair of points (z_i, z_j) separated by one dielectric resonant wavelength within the carbon nanotube, shifted from the left boundary of the structure by offsets specified in Fig. 8.14. (a) Circular dichroism $\Delta A = 0.8048 \mu\text{m}^{-1}$; (b) $\Delta A = 0.75392 \mu\text{m}^{-1}$; (c) $\Delta A = 0.6661 \mu\text{m}^{-1}$; (d) $\Delta A = 0.598 \mu\text{m}^{-1}$;

from level $|4''\rangle$ to $|2''\rangle$). The observed spectra in the latter case are reminiscent of electromagnetically-induced transparency (EIT) and coherent population trapping effects in a three-level system [51, 52]. In fact, the absorption at resonance is close to zero and the shape of the spectrum is similar to the absorption dip, observed in EIT. The predicted destructive interference in an external axial magnetic field after the passage of the ultrashort pulse is due to the specific timescales of the processes involved in the relaxation dynamics. This behaviour is confirmed by Fig. 8.19 where the induced phase shift is plotted as a function of wavelength. Whereas the phase shift spectrum for a σ^- pulse excitation is of the type of a two-level atomic phase shift, the shape of the phase shift curve is double-peaked which is characteristic for the real part of the susceptibility in a three-level system, exhibiting EIT. The calculated average specific rotatory power in a magnetic field ($B = 8$ T), is $-32.5804^\circ/\mu\text{m}$, corresponding to an average refractive index anisotropy of 6.497; the meaning of the minus sign is to denote left rotation (counterclockwise when looking against the light source). The predicted Faraday rotation at $B = 8$ T is nearly an order of magnitude greater than the natural optical rotation. We should note that the calculated rotation is a combined effect from the chirality of the nanotube and the magnetic field-induced rotation and thus can be considered as an estimate for the magneto-chiral effect in a single nanotube. Using the developed model investigation

of the magnetic field dependence of the optical rotation is currently under way and will be a subject of a further paper.

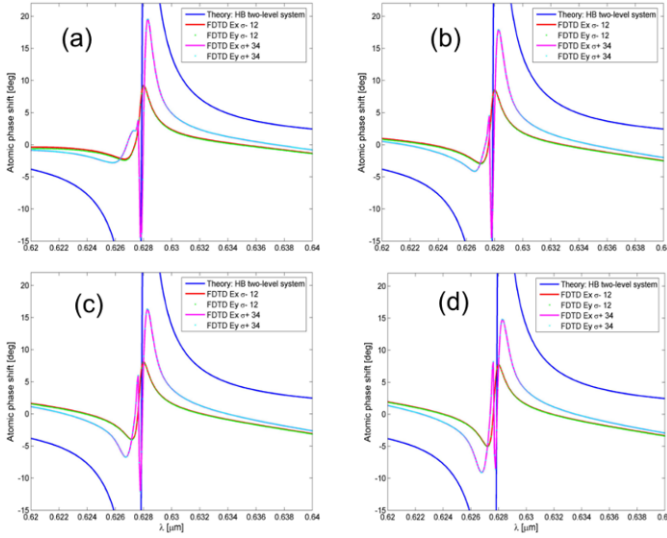


Fig. 8.19 Spatially resolved calculated phase shift vs wavelength at $B = 8$ T for E_x (red line) and E_y (green dot) components under σ^+ excitation and for E_x (magenta line) and E_y (cyan dot) under σ^- circularly polarised ultrashort optical excitation and the theoretical phase shift of a homogeneously broadened resonant two-level system (blue line) for the cases in Fig. 8.18: (a) $\Delta\phi = \beta l = 22.011^\circ$, $\rho = -40.4033^\circ/\mu\text{m}$; (b) $\Delta\phi = 21.378^\circ$, $\rho = -39.2413^\circ/\mu\text{m}$; (c) $\Delta\phi = 16.447^\circ$, $\rho = -30.19^\circ/\mu\text{m}$; (d) $\Delta\phi = 11.161^\circ$, $\rho = -20.4871^\circ/\mu\text{m}$;

8.6 Conclusions

We have developed a theoretical formalism and a dynamical model for description of the natural optical activity and Faraday rotation in an individual chiral SWCNT in the coherent nonlinear regime under resonant ultrashort polarised pulse excitation. The model is based on a discrete-level representation of the optically active states near the band edge, whereby chirality is modeled by four-level systems, specific for each handedness, that are mirror reflections of each other, and therefore non-superimposable. Thus chirality is incorporated at a microscopic level in the model. The dynamics of the resonant coherent interaction of a polarised ultrashort laser pulse with the discrete multilevel system are treated semiclassically within the coherent vector Maxwell-pseudospin formalism. For illustration purposes we consider the specific case of a (5,4) SWCNT, although the model is valid for an arbitrary chiral nanotube. Furthermore, we provide an estimate for the effective dielectric constant and the optical dipole matrix element for transitions excited by cir-

cularly polarised light in a single nanotube ((5,4) in particular). The model yields the time evolution of the optical pulse during its propagation due to the resonant coherent interactions, at each point along the nanotube and thus enables extracting the circular dichroism and phase shift spectra. Giant natural gyrotropy for the special case of a (5,4) nanotube considered ($3000^\circ/\text{mm}$ at $B = 0$), exceeding the one of birefringent crystals, liquid crystals and artificial metamaterials and thin films, is numerically demonstrated. Our results confirm the possibility to determine single nanotube handedness by time-resolved circular dichroism and magneto-optical rotatory dispersion spectroscopy.

In view of the possibility of engineering the nanotube structure, we anticipate much wider range of variation of the specific optical rotation. This is, however a subject of an ongoing further study using the proposed model and will not be discussed in this paper. We show that the circular dichroism and specific rotatory power in an axial magnetic field for the same (5,4) nanotube is enhanced at higher magnetic fields, e.g. $B = 8$ T. Up to our knowledge, the way the external axial magnetic field affects the natural optical activity and the so called magneto-chiral effect in the high-intensity nonlinear regime has not been investigated. These studies are currently under way, using the developed model. We should stress that although the model is developed for a resonant excitation, an off-resonant excitation, introducing detuning, can be easily incorporated.

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Chapter 9

Exciton and spin coherence in quantum dot lattices

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Abstract We theoretically investigate the optical properties of different semiconductor systems containing two dimensional lattices of neutral and charged quantum dots embedded in planar and arrays of optical cavities. The strong exciton (trion)-photon coupling is described in terms of polariton quasiparticles. First, we focus on a lattice of neutral dots in a planar microcavity. We show that Bragg polariton modes can be obtained by tuning the exciton and the cavity modes into resonance at high symmetry points of the Brillouin zone. The effective mass of these polaritons can be extremely small and makes of them the lightest exciton-like quasiparticles in solids. We analyze how disorder affects the properties of these Bragg polariton modes. It is found that in some cases weak disorder increases the light matter coupling and it leads to a larger polariton splitting. The second system investigated is similar to the first, but each dot has been charged with one electron. The electron spin determines the polarization of the cavity photon that couples to the dot. Such *spin lattice* can be used for quantum information processing and we show that a conditional phase shift gate with high fidelity can be obtained. Finally, we investigate exciton-photon quantum phase transitions in a planar lattice of one-mode cavities containing one neutral quantum dot each. Adopting the mean-field approximation we calculate exciton- and photon-phase diagrams and demonstrate that by controlling exciton- and photon-hopping energies a very rich scenario of coupled fermionic-bosonic quantum phase transitions appears.

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9.1 Introduction

In this chapter, we report on our theoretical investigations on artificial structures with confined photons and excitons having spatial periodicity comparable to their wavelength. The strong light-matter coupling between excitons and photons is often described in terms of polariton modes, which identify propagating electromagnetic modes in a medium with strong dispersive properties. Lately, there has been a considerable theoretical and experimental effort on polariton condensation in planar microcavities [1, 2, 3, 4]. In the simplest picture, the critical temperature for polariton condensation is inversely proportional to the polariton effective mass. Given their small effective mass (about four orders of magnitude smaller than excitons), quantum well polaritons suggest the possibility of creating a condensate at high temperature. Therefore, it is important to investigate diverse geometrical realizations of polaritonic structures, where the photon and matter excitations are confined in different ways. These diverse geometries could lead both to novel quantum information devices and to novel systems in which coherent matter states can be obtained at room temperature. The first system we will discuss is a lattice of quantum dots in a planar microcavity structure [5]. Here, a new idea consists in tuning the exciton energy in such a way that at least two photon modes with different momentum are resonantly coupled to it due to the lattice symmetry. There has been many proposals for engineering exciton-polaritons, or two level system-polaritons, using periodic structures: with periodic quantum well (QW) Bragg structures [6, 7], cavity-free three dimensional arrays of QDs [8] or two-level atoms [9], point-dipole crystal [10], photonic bandgaps with anti-dots [11], and confined QW polaritons in mesa structures [12]. Over all, two-dimensional polaritonic structures have the advantage that the in-plane momentum can be directly mapped onto an emission angle and dispersion law can be easily probed. In the proposed structure, the polariton dispersion is entirely determined by the coupling between the exciton in the dots and two dimensional photons, which leads to in-plane effective masses which can be exceptionally small, of order of $10^{-8}m_0$, for some highly symmetric points in the first Brillouin zone. The extremely small mass makes this structure a promising candidate for novel approaches to high temperature polariton condensation, and for long-range coupling of spin [13] or exciton [14] qubits. We also consider a lattice of charged quantum dots in planar photonic systems. In this case the polariton modes can lead to an effective interaction between electronic spins, and can have important applications for quantum computing implementations. We note that there has been a remarkable experimental progress towards these new systems as summarized in the recent review [15]. The last system we discuss consists of a two-dimensional array of cavities containing each one neutral quantum dot. Recently atoms in atomic lattices have been proposed as *quantum simulators* and used to investigate many body hamiltonians. This last semiconductor system can be seen as a semiconductor counterpart of those atomic systems, since fundamental many body effects with strong light-matter interaction can be investigated using this artificial structure. Since the dawn of quantum physics, there has been a long standing interest in the quantum phase transition (QPT), i.e. a phase change by changing an external parameter at *zero tem-*

perature driven by quantum fluctuations [16]. Recently, the QPT has attracted a lot of interest in many-body problems like strongly interacting electronic systems in condensed matter physics [17] or weakly interacting ultracold atomic systems [18]. Nevertheless, it is experimentally very demanding to observe such phenomena in these systems. It is somewhat easier in the case of optical lattices with ultracold atoms, allowing to conveniently study trapped Bose gases [19]. The system we are proposing is a new system which has not been investigated theoretically or realized experimentally so far. Our system can be realized as an array of QDs embedded in a photonic crystal, where the quantum dot is positioned on the site with a missing hole, i.e. zero dimensional cavity – nanocavity [20]. Although the experimental realization of the studied system might prove difficult in the near future, we believe that its theoretical investigation will shed light on two-component quantum phase transitions. The investigated model can be also regarded as a tight-binding approach for the study of the polariton condensation in a system of the quantum well embedded in a planar cavity [21]. Our approach allows to assess the influence of different exciton and photon effective masses via the introduction (and the variation over several orders of magnitude) of the exciton hopping t_X and the photon hopping t_P parameters. Thus, our model goes beyond the widely spread approach that always assumes photon coherence [4]. Consequently, it shows under which conditions the true polariton condensation can occur. This chapter is organized as follows. We first introduce in Section 9.2 the general formalism to describe our systems. Subsection 9.2.1 focuses on a lattice of neutral quantum dots in a planar cavity. We introduce directly the formalism that includes the effects of disorder, from which the zero-disorder case can be obtained as a limit. Subsection 9.2.2 describes the case of lattices of charged quantum dots in a cavity. We show how polariton modes can induce an effective spin hamiltonian for the localized electrons, which can be used in the realization of a quantum gate. Also in this subsection, we introduce the effects of cavity photon and exciton finite lifetimes and discuss the consequences for the fidelity of quantum operations. Finally, in Subsection 9.2.3 we introduce the Hamiltonian for the system composed of an array of cavities and quantum dots, and the mean field approach that will be used to study the different quantum phases. Numerical results are presented and discussed in Section 9.3. Subsections 9.3.1, 9.3.2, and 9.3.3 focus on the three different systems introduced in Section 9.2. Conclusions are in Section 9.4.

9.2 Theory

9.2.1 Neutral quantum dot lattice

We are going to investigate a disordered planar lattice of quantum dots embedded in a planar microcavity structure [5, 22]. We assume that only one excitonic level is present on each dot, implying strong localization. We start with the Hamiltonian in second quantization ($\hbar = 1$)

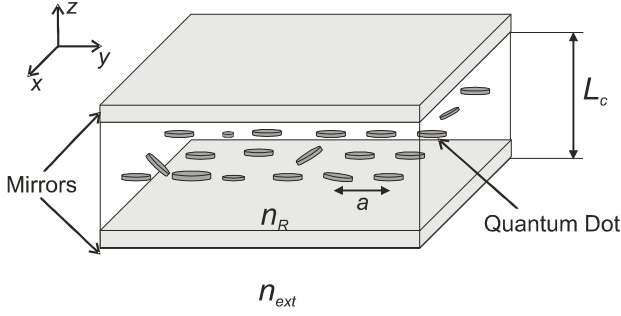


Fig. 9.1 Scheme of the investigated system. A lattice of quantum dots is embedded in a planar microcavity of length L_c and refractive index n_R (n_{ext} is an external refractive index). Energy, position, and oscillator strength of the localized excitons fluctuates from site to site.

$$\hat{H} = \sum_j \omega_j C_j^\dagger C_j + \sum_q \omega_q a_q^\dagger a_q + \sum_{jq} (ig_{jq} e^{iqR_j} a_q^\dagger C_j + h.c.) , \quad (9.1)$$

where $C_j^\dagger$ is the exciton creation operator on the j^{th} dot at position R_j with energy ω_j and exciton-photon coupling g_{jq} , $a_q^\dagger$ is the creation operator of the cavity photon mode with in-plane momentum q and energy ω_q . By writing the coupling constant as $g_{jq} e^{iqR_j}$ we can analyze separately the disorder effect induced by position fluctuations and oscillator strength fluctuations.

9.2.1.1 Ideal lattice

Assuming that all excitons have the same energy, light-matter coupling, and are found at the ideal positions the Hamiltonian Eq. (9.1) simplifies to

$$H = \omega_x \sum_j C_j^\dagger C_j + \sum_q \omega_q a_q^\dagger a_q + \sum_{j,q} \left\{ ie^{iqR_j} g a_q C_j^\dagger + h.c. \right\} , \quad (9.2)$$

where ω_x is the exciton energy and g the exciton-photon coupling constant at $R_j = 0$. The lattice symmetry can be exploited by introducing new exciton operators

$$C_q^\dagger = \frac{1}{\sqrt{N}} \sum_j C_j^\dagger e^{iqR_j} , \quad (9.3)$$

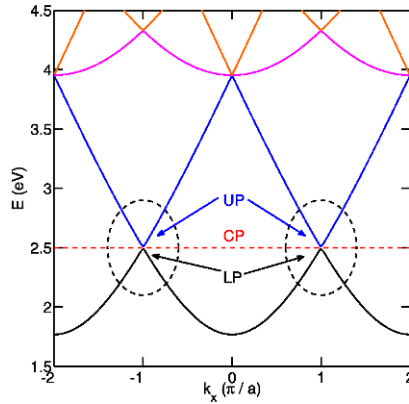
where N is the total number of lattice sites. This allows to rewrite the polariton Hamiltonian as

$$H = \sum_{q \in BZ} \left[\omega_x C_q^\dagger C_q + \sum_Q \omega_{q+Q} a_{q+Q}^\dagger a_{q+Q} + \sum_Q \left\{ g_{q+Q} a_{q+Q} C_q^\dagger + h.c. \right\} \right] , \quad (9.4)$$

where q is now restricted to the first Brillouin Zone (1st BZ) due to the periodicity of the lattice and Q is a reciprocal in-plane lattice vector. The renormalized coupling constant $g_q = \sqrt{N}g_{0,q}$ expressed in terms of materials and structure parameters is given in [5].

Moreover, the light-matter interaction conserves the momentum q only up to a reciprocal lattice vector. The terms in Eq. (9.4) involving the exchange of reciprocal lattice vectors are known as exciton *Umklapp*-processes in the literature [23, 24, 10]. This is in contrast to the QW microcavity case where there is a one-to-one correspondence between cavity and exciton modes (the in-plane momentum is conserved exactly). For an exciton state with momentum q away from the zone boundary,

Fig. 9.2 Scheme of photonic branches (solid lines) in a repeated zone scheme. The exciton energy band is indicated by the flat dashed line. Bragg polaritons are obtained by tuning the exciton energy at the zone boundary where photonic branches cross. Upper, Lower, and Central Polariton modes (indicated by UP, LP, and CP) appear due to the light-matter coupling.



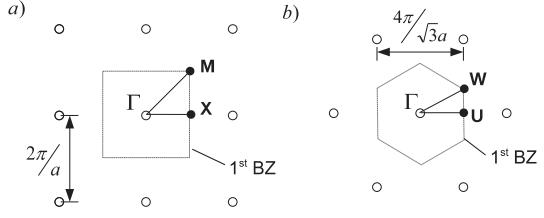
many off-resonant Umklapp terms give corrections which do not entail qualitative novel properties for the polariton quasi-particles. However, the situation can be very different if the structure is built in such a way that the exciton is resonant with the cavity at a q_0 which is at or near the BZ boundary. In that case we can choose, for instance, Q_i and Q_j so that the Bragg condition

$$\omega_x \sim \omega_{q_0-Q_i} \sim \omega_{q_0-Q_j} \quad (9.5)$$

is satisfied. In general, the number n of reciprocal lattice points satisfying this condition depends on the lattice symmetry and on the energy of the exciton. The mode configuration leading to the formation of Bragg cavity polaritons is illustrated in Fig. 9.2, where the exciton energy (red dashed horizontal line) is tuned in such a way that the polariton mixing occurs at the zone boundary. Due to the reduced symmetry of the dot lattice the $k_z = \pi/L_c$ cavity mode (see Fig. 9.1) is folded giving rise to many photonic branches, each characterized by a different reciprocal lattice vector Q . In order to visualize this folding, the figure shows two repeated BZs. Bragg polariton modes will appear at q_0 where two or more photonic branches are crossing and are nearly degenerate with the exciton energy. These polaritons will have different properties than polaritons usually investigated at $q_0 \sim 0$. Fig. 9.3 shows the high symmetry points at the zone boundary for the square and hexagonal lattice

for which we will discuss Bragg polaritons in the following. A cut-off in the cou-

Fig. 9.3 Reciprocal lattice of a square lattice (a) and of a hexagonal lattice with lattice constant a (b). High symmetry points discussed in the text are indicated.



pling constant g can be introduced due to the finite size of quantum dots. We will use the parameter β to characterize the dot size. The single polariton Hamiltonian at fixed q can be written in a matrix form [5]. Furthermore, the finite size of the dots limits the largest reciprocal lattice vector to $|Q_{max}| \sim 2\pi/\beta$. If we keep only the n resonant terms at the zone boundary satisfying exactly the Bragg condition, the polariton Hamiltonian can be diagonalized analytically and we find the two eigenvalues $\lambda_{1,2} = \omega_x \pm \sqrt{n}g_{q_0}$, corresponding to strongly mixed polariton states, as well as a $(n-1)$ -fold degenerate eigenvalue $\lambda_3 = \omega_x$. The normal mode splitting (Rabi energy), i. e. the difference between the lowest (lower polariton) and the highest (upper polariton) energy at resonance, is then

$$\Omega = 2\sqrt{n}g_{q_0}, \quad (9.6)$$

and is proportional to the square root of the number of reciprocal lattice vectors involved in the Bragg condition.

One of the most interesting features of Bragg polaritons consists in their extremely small effective mass, which is a consequence of the fact that two-dimensional photons at large momentum enter in the polariton formation. After a somewhat lengthy but straightforward calculation [5] we find that the effective mass ratio at X -point reads

$$\frac{m_{xx}}{m_{yy}} = \frac{\tilde{g}_X}{\sqrt{2}} \left(\frac{Ka}{\pi} \right)^2 \sim 10^{-3}. \quad (9.7)$$

where K is the photon momentum at X -point and a is the lattice constant. Therefore, the polariton mass is very anisotropic at the X -point. We can also compare the value of these effective masses with the QW polariton mass, which is of the same order of magnitude of the cavity photon mass at $q \sim 0$, $m_{ph} = \frac{\hbar n_R}{c} k_z$, where n_R is the refractive index and typically $\sim 10^{-5}m_0$. Then, we remark that for the the mass along the ΓX direction we have $m_{xx}/m_{ph} \sim 10^{-3}$ and $m_{yy}/m_{ph} \sim 1$. This implies that a value of the order of $10^{-8}m_0$ is expected. Moreover, we will see in the next section that isotropic masses of such a order can be obtained at special high symmetry points.

9.2.1.2 Disorder properties

We assume that the distance between dots is much larger than the effective exciton localization length on each dot. Therefore, fluctuations on different dots are uncorrelated. For energy fluctuations $\Delta\omega_j = \omega_j - \omega_X$ (ω_X is the average exciton energy), we have

$$\langle \Delta\omega_j \rangle = 0 \quad (9.8)$$

and

$$\langle \Delta\omega_m \Delta\omega_n \rangle = \sigma_\omega^2 \delta_{mn}, \quad (9.9)$$

where $\langle \cdot \rangle$ indicates averaging over a Gaussian ensemble. Similarly, for the oscillator strength disorder we have

$$\langle g_{jq} \rangle = \langle g_{0q} \rangle = \overline{g_{0q}}, \quad \langle g_{kq} g_{lq'} \rangle - \langle g_{kq} \rangle \langle g_{lq'} \rangle = \sigma_{qq'}^2 \delta_{kl}, \quad (9.10)$$

where $g_{0q} = g e^{-q^2 \beta^2/4}$, β is the characteristic exciton localization length[5], and

$$\sigma_{qq'}^2 = \sigma_g^2 e^{-(q^2 + q'^2) \beta^2/4}. \quad (9.11)$$

In Eqs. (9.10) and (9.12), $\sigma_g^2 = \langle g^2 \rangle - \langle g \rangle^2$ is the on-site oscillator strength variance at $q = 0$. Finally, for the positional disorder we obtain

$$\langle R_j \rangle = \overline{R_j}, \quad \langle R_k R_l \rangle - \langle R_k \rangle \langle R_l \rangle = \sigma_R^2 \delta_{kl}, \quad \langle e^{iqR_j} \rangle = e^{iq\overline{R_j}} e^{-q^2 \sigma_R^2/2}, \quad (9.12)$$

where the positions $\overline{R_j}$ identify the ideal two dimensional lattice. Furthermore, we assume that the different kinds of disorder are uncorrelated.

9.2.1.3 Polariton scattering

We can separate the disorder free part of the total Hamiltonian by defining the Fourier transform of the exciton operators as

$$C_j^\dagger = \frac{1}{\sqrt{N}} \sum_{q \in 1.BZ} e^{iq\overline{R_j}} C_q^\dagger, \quad C_j = \frac{1}{\sqrt{N}} \sum_{q \in 1.BZ} e^{-iq\overline{R_j}} C_q, \quad (9.13)$$

where $\overline{R_j}$ corresponds to the j^{th} site of a two dimensional ideal lattice and N is the total number of sites. The sum over the q states is restricted to the first Brillouin Zone (BZ) of the reciprocal lattice space. The disorder free Hamiltonian then takes the form of

$$\hat{H}_0 = \sum_q \left\{ \omega_X C_q^\dagger C_q + \sum_Q \omega_{q+Q} a_{q+Q}^\dagger a_{q+Q} + \sum_Q (\tilde{g}_{q+Q} a_{q+Q}^\dagger C_q + h.c.) \right\}, \quad (9.14)$$

where Q is a reciprocal lattice vector, and $\tilde{g}_q = \sqrt{N} g_{0q}$ is the renormalized coupling constant. We can then define the disorder coupling constants

$$\begin{aligned} \Delta \omega_q &= \frac{1}{\sqrt{N}} \sum_j e^{iq\bar{R}_j} \Delta \omega_j, \\ \eta_{qq'}^o &= \frac{1}{\sqrt{N}} \sum_j e^{i(q-q')\bar{R}_j} (g_{jq} - \bar{g}_{0q}), \\ \eta_{qq'}^p &= \frac{1}{\sqrt{N}} \sum_j (e^{iqR_j} - e^{iq\bar{R}_j}) e^{-iq'\bar{R}_j} \bar{g}_{0q}, \end{aligned} \quad (9.15)$$

where the index o (p) indicates oscillator strength (positional) disorder. The statistical properties of these functions are determined by their average values as

$$\langle \Delta \omega_k \rangle = 0, \quad \langle \eta_{qq'}^o \rangle = 0, \quad \langle \eta_{qq'}^p \rangle = \sum_K \delta_{q,q'+K} \xi_q, \quad (9.16)$$

and correlations according to

$$\begin{aligned} \langle \Delta \omega_k \Delta \omega_l^* \rangle &= \sigma_\omega^2 \delta_{kl}, \\ \langle \eta_{qq'}^o \eta_{kk'}^{*o} \rangle &= \sum_K \delta_{q-k, q'-k'+K} \sigma_{qk}, \\ \langle \eta_{qq'}^p \eta_{kk'}^{*p} \rangle - \langle \eta_{qq'}^p \rangle \langle \eta_{kk'}^{*p} \rangle &= \sum_K \delta_{q-k, q'-k'+K} \zeta_{qk}, \end{aligned} \quad (9.17)$$

where K is a reciprocal lattice vector, and

$$\begin{aligned} \xi_q &= \tilde{g}_{0q} (e^{-q^2 \sigma_R^2/2} - 1), \\ \zeta_{qk} &= \bar{g}_{0q} \bar{g}_{0k}^* (e^{-|q+k|^2 \sigma_R^2/2} - e^{-q^2 \sigma_R^2/2} e^{-k^2 \sigma_R^2/2}). \end{aligned} \quad (9.18)$$

The disorder terms of the Hamiltonian for the three different mechanisms (energy, oscillator strength, and position) can then be written in a compact form as

$$\hat{H}^e = \frac{1}{\sqrt{N}} \sum_{qk} \Delta \omega_k C_{q+k}^\dagger C_q, \quad \hat{H}^{o(p)} = \sum_{qq'} (\eta_{qq'}^{o(p)} a_q^\dagger C_{q'} + h.c.). \quad (9.19)$$

We can find the eigenvalues and eigenvectors of the disorder free Hamiltonian by solving the problem

$$\hat{H}_0 |P_{nq}\rangle = \Lambda_{nq} |P_{nq}\rangle. \quad (9.20)$$

The corresponding eigenstates are the disorder-free polariton states, which can be written in the form

$$|P_{nq}\rangle = P_{nq}^\dagger |0\rangle = (u_{nq}C_q^\dagger + \sum_Q v_{nq+Q}a_{q+Q}^\dagger)|0\rangle, \quad (9.21)$$

where n is a band index, v_{nq} and u_{nq} are Hopfield coefficients[25], and $|0\rangle$ is the exciton-photon vacuum. We can use these states and obtain an effective disorder potential for polaritons as

$$V_{nn'qq'}^J = \langle P_{nq} | \hat{H}_{1(2)} | P_{n'q'} \rangle,$$

where $J \in \{e, o, p\}$ labels energy, oscillator strength, and positional disorder, respectively. This gives for the energy disorder

$$V_{nn'qq'}^e = \frac{\omega_{q-q'}}{\sqrt{N}} u_{nq}^* u_{n'q'}, \quad (9.22)$$

and for the oscillator strength (positional) disorder

$$V_{nn'qq'}^{o(p)} = \sum_Q \left(\eta_{q+Qq'}^{o(p)} u_{n'q'}^* v_{nq+Q}^* + \eta_{q+Qq'}^{*o(p)} u_{nq}^* v_{n'q'+Q}^* \right). \quad (9.23)$$

9.2.1.4 Absorption spectrum

The absorption spectrum at an angle determined by the inplane q of the cavity photon can be calculated using the full propagator of the disordered system, which can be written in terms of the disorder-free polariton states as

$$\hat{G}(\omega) = \sum_{nn'kk'} |P_{nk}\rangle G_{nn'kk'}(\omega) \langle P_{n'k'}| \quad (9.24)$$

with

$$G_{nn'kk'}(\omega) = G_{nk}^0(\omega) \delta_{nn'} \delta_{kk'} + G_{nk}^0(\omega) T_{nn'kk'}(\omega) G_{n'k'}^0(\omega), \quad (9.25)$$

where $G_{nk}^0(\omega) = \frac{1}{\omega - \Lambda_{nk} + i\epsilon}$ is the Green's function for disorder-free polaritons. T is the scattering T-matrix that can be expressed as

$$T(\omega) = \sum_j T^{(j)}(\omega),$$

$$T_{nn'kk'}^{(j)}(\omega) = \langle P_{nk} | \hat{V} (\hat{G}^0(\omega) \hat{V})^j | P_{n'k'} \rangle. \quad (9.26)$$

The imaginary part of the polariton propagator projected on a photon mode at a given wavevector $|\gamma\rangle \equiv |k_\gamma\rangle$ gives the absorption spectrum at the corresponding excitation angle as

$$A_\gamma(\omega) = -\text{Im} G_\gamma(\omega), \quad (9.27)$$

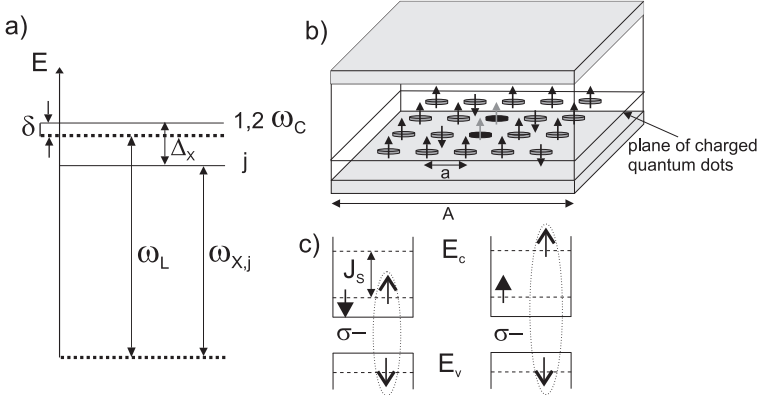


Fig. 9.4 (a) Energy diagram with cavity energy ω_C , exciton energy $\omega_{X,j}$ and detuning Δ_X (see text for details), and laser frequency ω_L . (b) Scheme of the spin lattice composed by charged quantum dots in a planar cavity. Two dots brought into resonance with the cavity are highlighted. (c) Diagram of allowed spin configurations for a charged dot excited by circularly polarized light. The distance between the trion energy of the two configurations defines an antiferromagnetic spin coupling between the electron spin and the exciton spin (polarization).

with

$$G_\gamma(\omega) = \langle \gamma | \hat{G}(\omega) | \gamma \rangle = \sum_{nn'} v_{nk_\gamma} v_{n'k_\gamma}^* G_{nn'k_\gamma k_\gamma}(\omega).$$

We can explicitly average over the disorder configurations to obtain

$$\langle G_\gamma(\omega) \rangle = \sum_{nn'} v_{nk_\gamma} v_{n'k_\gamma}^* \langle G_{nn'k_\gamma k_\gamma}(\omega) \rangle$$

with

$$\langle G_{nn'k_\gamma k_\gamma}(\omega) \rangle = G_{nk}^0(\omega) \delta_{nn'} \delta_{kk'} + G_{nk_\gamma}^0(\omega) \langle T_{nn'k_\gamma k_\gamma}(\omega) \rangle G_{n'k_\gamma}^0(\omega). \quad (9.28)$$

In the case of disorder caused by energy inhomogeneity, the ensemble averaging leads to the momentum conservation, $\langle T_{nn'kk'}(\omega) \rangle = \langle T_{nn'kk}(\omega) \rangle \delta_{kk'}$, since by averaging only the diagonal terms in Eq. (9.17) remain. We note that this is similar to the problem of a single particle scattering in a random ensemble of impurities [26]. The diagonal elements of the T -matrix can be interpreted as an effective self-energy, $\Sigma_{nk}(\omega) = T_{nnkk}(\omega)$, and within the pole approximation $\Sigma_{nk}(\omega = \Lambda_{nk})$ their real part gives an energy shift of the polariton levels.

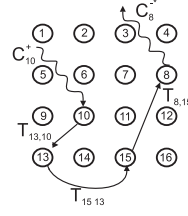
9.2.2 Charged quantum dot lattice

Our assumptions for the system studied in this subsection (see Fig.9.4b) are the following [27]: (i) the trion energy $\omega_{X,j}$ of each dot can be independently controlled e.g. by applying a local voltage [28], (ii) the quantum dots are well separated so there is not direct overlap of the trion wavefunction, (iii) each dot can be occupied only by one additional exciton, (iv) the heavy-hole light-hole splitting is large enough that only the heavy-hole exciton is taken into account, and (v) the cavity and the spin lattice are ideal. The role of the cavity is to enhance the range of the interaction between dots [14] and their spins [13]. The long-range coupling is mainly mediated by polariton modes with in-plane wavevectors $q \sim 0$, therefore in the following we neglect cavity modes with $q \neq 0$. The total Hamiltonian describing the spin lattice consists of the three terms: a non-Hermitian [29] polariton term $\hat{H}_P$ describing the interaction of the exciton with the cavity mode, an exciton-spin term $\hat{H}_I$, and a cavity-laser term $\hat{H}_L$ describing the pumping of the cavity mode by an external laser, which is described using the quasi-mode model [21]. These terms can be written as

$$\begin{aligned}\hat{H}_P &= \sum_{\sigma} \left\{ - \sum_j (\omega_{X,j} - i\Gamma) C_{j\sigma}^{\dagger} C_{j\sigma} + g \sum_j (a_{\sigma} C_{j\sigma}^{\dagger} + h.c.) + (\omega_C - i\kappa) a_{\sigma}^{\dagger} a_{\sigma} \right\}, \\ \hat{H}_I &= \sum_j J_S S_{jz} P_{jz}, \quad \hat{H}_L = \sum_{\sigma} (V_{\sigma} e^{i\omega_L t} a_{\sigma} + h.c.),\end{aligned}\tag{9.29}$$

where $C_{j\sigma}^{\dagger}$ ($C_{j\sigma}$) is the creation (annihilation) operator of exciton on the j^{th} dot at position R_j with polarization σ and decay rate Γ , $a_{\sigma}^{\dagger}$ (a_{σ}) is the creation (annihilation) operator of the cavity photon with the energy ω_C [5] and cavity leakage rate κ , g is the dot-photon coupling constant. The exciton-spin coupling constant in $\hat{H}_I$, J_S , is defined as the energy difference between trion states with parallel and anti-parallel spins as schematically shown in Fig. 9.4c; S_{jz} is the z -component of the electron spin in the j^{th} QD; and $P_{jz} = C_{j\uparrow}^{\dagger} C_{j\uparrow} - C_{j\downarrow}^{\dagger} C_{j\downarrow}$ is the operator corresponding to the z component of the exciton polarization. In $\hat{H}_L$, V_{σ} is the laser-cavity coupling constant and ω_L is the frequency of the external laser. A $\sigma+$ ($\sigma-$) polarized photon creates a bright exciton with $\downarrow$ ($\uparrow$) electron spin in the growth (z) direction. For excitons in III-V confined systems the possible values of the electron spin are $\sigma_z^e = \pm \frac{1}{2}$ and the heavy hole spin are $\sigma_z^{hh} = \mp \frac{3}{2}$. The $\sigma+$ ($\sigma-$) circularly polarized light leads to an effective magnetic field (and higher order odd terms) in the positive (negative) z direction with strength proportional to the light intensity [30]. We assume throughout this section that the light is linearly polarized, which makes all multi-spin terms of odd order identically zero. This is caused by the fact that these terms would break the time-reversal symmetry [31], which has to be preserved in the presence of linearly polarized radiation. We remark that our method is not restricted to the linearly polarized light. The usage of other polarizations would complicate the Hamiltonian but would not change the main conclusions.

Fig. 9.5 Diagram illustrating multiple scattering events that lead to a multi-spin coupling $J_{8,15,13,10}^{(4)}$ as derived in Eq. (9.35).



9.2.2.1 Multi-Spin Hamiltonian

The effective spin Hamiltonian can be calculated introducing the level shift operator $R(\omega_L)$ in [32]

$$\hat{H}_s = \mathcal{P}R(\omega_L)\mathcal{P} = \mathcal{P}\hat{H}_L \frac{\mathcal{Q}}{\omega_L - \mathcal{Q}(\hat{H}_P + \hat{H}_I)\mathcal{Q}} \hat{H}_L \mathcal{P}, \quad (9.30)$$

where $\mathcal{P} = \sum_{\lambda} |\lambda\rangle\langle\lambda| \otimes |0\rangle\langle 0|$ is the projection operator on the subspace of all spin states λ and no polaritons and $\mathcal{Q} = 1 - \mathcal{P}$. Using the rotating wave approximation and assuming linearly polarized laser, the cavity-laser term can be rewritten as $\hat{H}_L = V_{\downarrow}a_{\downarrow} + V_{\uparrow}a_{\uparrow} + h.c.$. We solve first the polariton problem for both polarizations and obtain the polariton states $|\nu \uparrow(\downarrow)\rangle$ satisfying $\hat{H}_P|\nu \uparrow(\downarrow)\rangle = \omega_{\nu}|\nu \uparrow(\downarrow)\rangle$. The polariton states can be written in terms of excitons and cavity photon as $|\nu \uparrow(\downarrow)\rangle = (\sum_j u_{\nu j} C_{j\uparrow(\downarrow)}^{\dagger} + v_{\nu} a_{\uparrow(\downarrow)}^{\dagger})|0\rangle$. The coefficients Hopfield $u_{\nu j}$ represent the projection of the polariton state ν on the exciton state localized at the j^{th} dot. Similarly, v_{ν} represents the projection of the polariton state on the cavity photon. Using these coefficients the Hamiltonian $\hat{H}_s$ projected on the spin basis $|\lambda\rangle$ reads

$$H_s^{\lambda\lambda'} = \sum_{\mu\nu} v_{\mu} v_{\nu}^* \sum_{\sigma=\uparrow,\downarrow} \frac{V_{\sigma}^2}{2} \langle \mu\sigma | \langle \lambda | \{ \omega_L - (\hat{H}_P + \hat{H}_I) \}^{-1} | \lambda' \rangle | \nu\sigma \rangle. \quad (9.31)$$

The off-diagonal terms $\langle \lambda | \{ \omega_L - (\hat{H}_P + \hat{H}_I) \}^{-1} | \lambda' \rangle$ are zero since all spin dependent terms are proportional to S_z . This allows us to calculate the eigenenergies of the Hamiltonian $\hat{H}_s$ Eq. (9.31) exactly by inversion of the matrix. Perturbation theory can also be applied by expanding the resolvent as

$$\begin{aligned} \frac{1}{\omega_L - (\hat{H}_P + \hat{H}_I)} &= \frac{1}{\omega_L - \hat{H}_P} + \frac{1}{\omega_L - \hat{H}_P} \hat{H}_I \frac{1}{\omega_L - \hat{H}_P} \\ &+ \frac{1}{\omega_L - \hat{H}_P} \hat{H}_I \frac{1}{\omega_L - \hat{H}_P} \hat{H}_I \frac{1}{\omega_L - \hat{H}_P} + \dots \end{aligned} \quad (9.32)$$

Light with circular $\sigma+$ ($\sigma-$) polarization contributes with terms $\hat{H}_I \sim J_S$ ($\sim -J_S$) according to Eq. (9.29), consequently the odd terms, $\sim J_S^{2n+1}$, in Eq. (9.32) cancel out for linearly polarized light. After some straightforward algebra we can rewrite the effective spin Hamiltonian as

$$\hat{H}_s = \tilde{J}^{(0)} + \sum_{i>j} \tilde{J}_{ij}^{(2)} S_{iz} S_{jz} + \sum_{i>j>k>l} \tilde{J}_{ijkl}^{(4)} S_{iz} S_{jz} S_{kz} S_{lz} + \dots, \quad (9.33)$$

where the coupling constants are renormalized to take into account multiple scattering, e.g.

$$\tilde{J}_{12}^{(2)} = J_{12}^{(2)} + J_{21}^{(2)} + \sum_{iP} J_{P(12ii)}^{(4)} + \sum_{ijP} J_{P(12iijj)}^{(6)} + \dots, \quad (9.34)$$

where P indicates a permutation of all the indices. The multi-spin coupling constants $J_{i_1 \dots i_n}^{(n)}$ can be expressed as

$$J_{i_1 \dots i_n}^{(n)} = J_S^n V_{LP}^2 (C_{i_1}^-)^* T_{i_1 i_2} \dots T_{i_{n-1} i_n} C_{i_n}^+ \quad (9.35)$$

in terms of the photon-exciton coupling function $C_i^{+(-)}$ and exciton inter-dot transfer amplitudes T_{ij} defined as (see scheme in Fig. 9.5)

$$C_i^{+(-)} = \sum_{\mu} \frac{v_{\mu} u_{\mu i}^*}{\omega_L - \omega_{\mu}}, \quad T_{ij} = \sum_{\mu} \frac{u_{\mu i} u_{\mu j}^*}{\omega_L - \omega_{\mu}}, \quad (9.36)$$

where $V_{LP}^2 = \frac{V_{\uparrow}^2 + V_{\downarrow}^2}{2}$ is the effective light-polariton coupling constant. Note that due to the non-Hermitian nature of the polariton Hamiltonian the polariton energies ω_{μ} are complex quantities with the imaginary part $\gamma_{\mu} = \text{Im } \omega_{\mu}$ representing the polariton linewidth.

9.2.2.2 Fidelity of a conditional phase shift gate

Let us now consider two dots labeled by $\{1, 2\}$ in resonance with the lowest cavity mode, i.e. $\omega_{X,1(2)} = \omega_C$. The laser is detuned below the cavity mode as $\omega_L = \omega_C - \delta$, where δ is the *laser detuning*. The remaining quantum dots are detuned : $\omega_{X,1(2)} - \omega_{X,j \neq 1,2} = \Delta_X$, where Δ_X is the *exciton detuning* (schematically shown in Fig. 9.4 (a)).

We want to use the quantum dots $\{1, 2\}$ to test the feasibility of a conditional phase gate. The conditional phase gate (PG), is a universal two-qubit gate, i.e. it can realize universal quantum computation when combined with single qubit operations [33]. The gedanken gate sequence can be described as follows: (i) single qubit operations are performed on the two selected dots $\{1, 2\}$ (e.g. by optical control [34, 35]) then (ii) they are brought adiabatically into resonance with the cavity by controlling the exciton energy with local electric field, (iii) the laser is switched on adiabatically for a time t_C , and (iv) dots are brought back into the off-resonant state. We can estimate the error in the implementation of a conditional phase shift gate due to multi-spin interaction terms. A quantitative measure of the gate quality can be given using the gate *fidelity* [36] defined as $\mathcal{F} = |\langle \Psi | U_I^{\dagger} U_R | \Psi \rangle|^2$, where U_I

is the ideal gate matrix, and U_R is the real gate matrix, i.e. the one that includes the effects of multi-spin terms. Ψ is an arbitrary initial pure state, and $|\langle \Psi | \cdot | \Psi \rangle|^2$ indicates averaging over all pure initial states. Working in the basis of the full spin-Hamiltonian eigenstates $\{\phi_i\}$ (with 2^{N_D} states where N_D is dots number density), we can define an eigenvector fidelity as $\mathcal{F}_i = \langle \phi_i | U_I^\dagger U_R | \phi_i \rangle$. In order to calculate the fidelity, we calculate the time evolution operator $U(t_C) = \exp\{-iH_S t_C\}$ with a time $t_C = \frac{\pi}{2|J_{12}^{(2)}|}$ to obtain maximum fidelity. Besides multi-spin effects, the computation can be also spoiled by the exciton decay or by photon leakage. In order to estimate the effect of the exciton spontaneous emission and the photon leakage in the spin-spin coupling we consider the first two terms in Eq. (9.33) and compare the real part of the second term, which gives the effective spin-spin interaction $J_{12}^{(2)}$, and the imaginary part of the first term, which gives the decay rate independent of the spin state. The latter describes the process in which the polariton leaves the cavity without any interaction with the spins, which would fail the gate operation (even if it does not spoil the spin coherence). Introducing for simplicity a constant polariton linewidth γ we obtain

$$\text{Re}J_{12}^{(2)} \sim \frac{V_{LP}^2 J_S^2 |v|^2 |u|^2}{(\delta^2 + \gamma^2)^3} (\delta^3 - 3\delta\gamma^2), \quad \text{Im}J^{(0)} \sim -\frac{V_{LP}^2 |v|^2}{(\delta^2 + \gamma^2)} \gamma. \quad (9.37)$$

where $|v|^2$ and $|u|^2$ are the photon and the exciton part of the polariton. Note that in the limit $g \rightarrow 0$ we have $u \rightarrow 0$ and $v \rightarrow 1$, i.e. the coupling of the cavity photon and the exciton is necessary for the spin coupling. Together with the condition to have a small probability of the polariton emission during the gate operation, $\frac{\text{Im}J^{(n)}}{\text{Re}J^{(n)}} \sim \frac{\gamma}{\delta} \ll 1$ in the limit of $\delta \gg \gamma$, this translates into

$$\frac{\text{Im}J^{(0)}}{\text{Re}J_{12}^{(2)}} \sim \frac{\gamma\delta}{J_S^2} \ll 1, \quad (9.38)$$

which requires $J_S \gg \delta$. Using a quantum jump approach [37], the nonzero probability of state decay can be calculated as $P_D = 1 - P_{ND} = 1 - \frac{1}{N_D} \text{tr}\{|U(t_C)|\}$, where P_{ND} is a probability of a non-decayed state. Assuming that there has not been any decay, the $U(t_C)$ is then renormalized to one for the calculation of the fidelity [29].

9.2.3 Neutral quantum dots in lattice of optical cavities

Finally in this section, we investigate a system consisting of a two-dimensional array of one-mode polarization-selective cavities containing one quantum dot (QD) each as schematically shown in Fig. 9.6 and detailed in [38]. In order to concentrate on the most important aspects of the system we do not detail the precise material structure but rather write down our assumptions: (i) A strong exciton localization, which guarantees that at most two excitons with opposite spins can be found in each

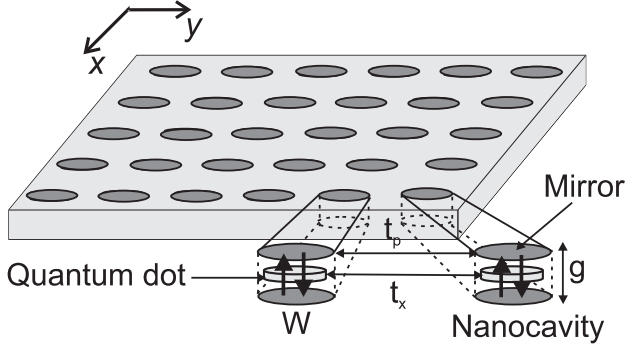


Fig. 9.6 Scheme of the investigated system: an array of the coupled nanocavity quantum dots embedded in a photonic bandgap structure. Photon (exciton) hopping energy t_p (t_x), light-matter coupling g , and exciton-exciton interaction W are indicated.

quantum dot. The term exciton spin is used for the z (growth direction)-component of the total exciton angular momentum. There are four possibilities, two optically active (bright) excitons with spin ± 1 and two optically nonactive (dark) excitons with spin ± 2 . In the following we restrict our discussion only to the bright ones. (ii) A spin-independent exciton energy ω_X . (iii) Existence of exciton transfer without specifying its dominant nature, i.e. Förster transfer [39] for short distances or radiative coupling [40, 14] for long distances, and additionally we assume that the most important transfer is the one between the nearest neighbors. (iv) Existence of photon hopping among nanocavities (photonic bandgap structure), as recently discussed [20, 41]. The Hamiltonian in the second quantization with exciton $\hat{H}_X$, photon $\hat{H}_P$, and exciton-photon $\hat{H}_{XP}$ parts therefore reads

$$\begin{aligned}\hat{H}_X^{(1)} &= \omega_X \sum_{j,\alpha} C_{j\alpha}^\dagger C_{j\alpha} - t_X \sum_{\langle j,k \rangle \alpha} C_{j,\alpha}^\dagger C_{k,\alpha} + \sum_j W \sum_j C_{j\uparrow}^\dagger C_{j\uparrow} C_{j\downarrow}^\dagger C_{j\downarrow} \\ \hat{H}_P^{(1)} &= \omega_C \sum_{j\alpha} a_{j\alpha}^\dagger a_{j\alpha} - t_P \sum_{\langle j,k \rangle \alpha} a_{j,\alpha}^\dagger a_{k,\alpha} \quad \hat{H}_{XP}^{(1)} = g \sum_{j\alpha} (a_{j\alpha}^\dagger C_{j\alpha} + a_{j\alpha} C_{j\alpha}^\dagger) \\ \hat{H} &= \hat{H}_X^{(1)} + \hat{H}_P^{(1)} + \hat{H}_{XP}^{(1)} - \mu N\end{aligned}\tag{9.39}$$

where $C_{j\alpha}^\dagger$ is the exciton fermionic creation operator on the j^{th} site with electron spin projection $\alpha = \uparrow, \downarrow$, t_X (t_P) is the exciton (photon) inter-site energy transfer between the nearest sites $\langle j, k \rangle$, $a_{j\alpha}^\dagger$ is the bosonic creation operator of the j^{th} nanocavity photon mode with circular polarization α and energy ω_C , g is the light-matter coupling, W is the exciton-exciton interaction potential, μ is the chemical potential, $N = \sum_{j\alpha} (C_{j\alpha}^\dagger C_{j\alpha} + a_{j\alpha}^\dagger a_{j\alpha})$ is the total number of particles. The on-site nanocavity-laser coupling can be described in the quasimode approximation as $\hat{H}_j^{CL} = \Omega_\uparrow a_{j\uparrow}^\dagger + \Omega_\downarrow a_{j\downarrow}^\dagger$ with Ω_α being the coupling constant. Exciting the nanocavity with linearly polarized light $\Omega_\uparrow = \Omega_\downarrow$ and rotating the basis to $a_{jX} = \frac{1}{\sqrt{2}}(a_{j\uparrow} + a_{j\downarrow})$ and $a_{jY} = \frac{1}{\sqrt{2}}(a_{j\uparrow} - a_{j\downarrow})$, modifies the exciton-photon coupling

to $\sim a_{jX}^\dagger (C_{j\uparrow} + C_{j\downarrow})$ and $\sim a_{jY}^\dagger (C_{j\uparrow} - C_{j\downarrow})$. Furthermore, we make a new assumption of a polarization-selective cavity, which is nontrivial from an experimental point of view. Consequently, assuming an X -polarization nanocavity all terms $\sim a_{jY}$ can be neglected since the exciton states couple either to X or Y polarization [X (Y) exciton $C_{X(Y)}^\dagger = \frac{1}{\sqrt{2}}(C_{\uparrow}^\dagger \pm C_{\downarrow}^\dagger)$]. Moreover, strong exciton spin-flip processes due to scattering are assumed to guarantee the same chemical potential for both spin up and spin down excitons. Finally, the following effective Hamiltonian is obtained

$$\begin{aligned}\hat{H}_P^{(2)} &= \omega_C \sum_j a_j^\dagger a_j - t_P \sum_{\langle j,k \rangle} a_j^\dagger a_k \quad \hat{H}_{XP}^{(2)} = g \sum_{j\alpha} (a_j^\dagger C_{j\alpha} + a_j C_{j\alpha}^\dagger) \\ \hat{H} &= \hat{H}_X^{(1)} + \hat{H}_P^{(2)} + \hat{H}_{XP}^{(2)} - \mu N\end{aligned}\quad (9.40)$$

where the operator of the number of particles changes to $N = \sum_{j\alpha} C_{j\alpha}^\dagger C_{j\alpha} + \sum_j a_j^\dagger a_j$. The sign and the magnitude of the exciton-exciton interaction potential W depend on the details of electron and hole confinement in the QD [42, 43, 44]. The potential W can be tuned e.g. by static electric field along z -direction or by applying a perpendicular magnetic field. Assuming that at zero magnetic field, there is only a biexciton binding energy $W = E_{XX} < 0$, then for stronger magnetic field, potential W changes by the effective exciton Zeeman splitting [45], $W = E_{XX} + \mu_B g_X B_z$, where μ_B is the Bohr magneton, g_X is the exciton g -factor, and B_z stands for the perpendicular magnetic field.

A close inspection of the Hamiltonian Eq. (9.40) furthermore reveals that it comprises two well-known Hamiltonians: (i) The Hubbard Hamiltonian [26]

$$\hat{H}^H = \omega_X \sum_{j\alpha} C_{j\alpha}^\dagger C_{j\alpha} - t_X \sum_{\langle j,k \rangle \alpha} C_{j\alpha}^\dagger C_{k\alpha} + W \sum_j C_{j\uparrow}^\dagger C_{j\uparrow} C_{j\downarrow}^\dagger C_{j\downarrow}, \quad (9.41)$$

for which insulator-metal transitions have been intensively studied, and (ii) the Jaynes-Cummings Hamiltonian

$$\hat{H}_{j\alpha}^{JC} = \omega_X C_{j\alpha}^\dagger C_{j\alpha} + \omega_C a_j^\dagger a_j + g(a_j^\dagger C_{j\alpha} + a_j C_{j\alpha}^\dagger), \quad (9.42)$$

which has received a lot of attention recently.

In order to study the phase transition of model Eq. (9.40), we stay within the meanfield theory, which generally gives a very good description in accordance with the Monte Carlo simulation [46]. In analogy to classical phase transitions or Bogolyubov approach and the idea of (spontaneous) symmetry breaking, we introduce the *exciton superfluid order parameter* $\psi_\alpha = \langle C_{i\alpha}^\dagger \rangle$ and the *photon coherence parameter* $\chi = \langle a_i^\dagger \rangle$. Although, we note that the meanfield approach generally works the better the higher the dimensionality of the system is, a two-dimensional system is still acceptable [16]. Thus, adopting the decoupling approximation

$$\begin{aligned}C_{i\alpha}^\dagger C_{j\alpha} &= C_{i\alpha}^\dagger \langle C_{j\alpha} \rangle + \langle C_{i\alpha}^\dagger \rangle C_{j\alpha} - \langle C_{i\alpha}^\dagger \rangle \langle C_{j\alpha} \rangle \\ a_i^\dagger a_j &= a_i^\dagger \langle a_j \rangle + \langle a_i^\dagger \rangle a_j - \langle a_i^\dagger \rangle \langle a_j \rangle,\end{aligned}\quad (9.43)$$

where i is not equal to j , the Hamiltonian Eq. (9.40) in the meanfield approximation reads

$$\begin{aligned}\hat{H}_X^{(2)} &= \sum_j \left(\omega_X \sum_{\alpha} C_{j\alpha}^{\dagger} C_{j\alpha} + W C_{j\uparrow}^{\dagger} C_{j\uparrow} C_{j\downarrow}^{\dagger} C_{j\downarrow} - z t_X \left[\sum_{\alpha} (C_{j\alpha}^{\dagger} \psi_{\alpha} + C_{j\alpha} \psi_{\alpha}^* - |\psi_{\alpha}|^2) \right] \right) \\ \hat{H}_P^{(3)} &= \sum_j \left(\omega_C a_j^{\dagger} a_j - z t_P [(a_j^{\dagger} \chi + a_j \chi^*) - |\chi|^2] \right) \\ \hat{H}_{MF} &= \hat{H}_X^{(2)} + \hat{H}_P^{(3)} + \hat{H}_{XP}^{(2)} - \mu N\end{aligned}\quad (9.44)$$

where z is the number of the nearest neighbor dots. Another simplification is possible since we have performed test calculations for complex $\psi_{\uparrow}$, $\psi_{\downarrow}$, and χ which have shown that (i) $\psi_{\uparrow}$ and $\psi_{\downarrow}$ are identical and (ii) *phase locking* between both parameters $\psi \equiv \psi_{\uparrow} = \psi_{\downarrow}$ and χ exists, which also follows from the symmetry of the Hamiltonian. This means that e.g. in the case of energy it holds $E(\psi, \chi) = E(\psi e^{i\beta}, \chi e^{i\beta})$, where β is an arbitrary real number. Recent studies of exciton phase transitions in coupled quantum wells have shown that depending on the sign of the exciton-exciton interaction W , the exciton ground state can be either paramagnetic ($\psi_{\uparrow} = \psi_{\downarrow}$ for $W < 0$) or ferromagnetic ($\psi_{\uparrow} \neq 0$ and $\psi_{\downarrow} = 0$ for $W > 0$) [42, 43]. However, in the present case the light-matter coupling g plays a very important role tending to equalize spin populations if it is sufficiently strong ($g \sim W$). It implies the convenient usage of a spin independent order parameter $\psi_{\uparrow} = \psi_{\downarrow} = \psi$. Furthermore, we note that the reality of ψ and χ is a well-established property of the meanfield [47] if they are independent. In the present case, they are coupled only indirectly via the light-matter coupling $\sim g$, which additionally explains the phase locking. Taking into account the above, the meanfield Hamiltonian Eq. (9.44) is modified as

$$\begin{aligned}\hat{H}_X^{(3)} &= \sum_j \left(\omega_X \sum_{\alpha} C_{j\alpha}^{\dagger} C_{j\alpha} + W C_{j\uparrow}^{\dagger} C_{j\uparrow} C_{j\downarrow}^{\dagger} C_{j\downarrow} - z t_X \left[\sum_{\alpha} (C_{j\alpha}^{\dagger} + C_{j\alpha}) \psi - 2|\psi|^2 \right] \right) \\ \hat{H}_P^{(4)} &= \sum_j \left(\omega_C a_j^{\dagger} a_j - z t_P [(a_j^{\dagger} + a_j) \chi - |\chi|^2] \right) \\ \hat{H}_{MF} &= \hat{H}_X^{(3)} + \hat{H}_P^{(4)} + \hat{H}_{XP}^{(2)} - \mu N\end{aligned}\quad (9.45)$$

We note that if $\chi \neq 0$ photons are in a coherent state and if $\chi = 0$ they are in a Fock state. For convenience, the exciton-photon *detuning* can be defined as

$$\Delta = \omega_X - \omega_C. \quad (9.46)$$

The case without exciton and photon transfers ($t_X = 0$ and $t_P = 0$), i.e. exciton and photon insulator, can be solved easily. One only needs to diagonalize the Hamiltonian matrix, which in the basis of n excitations $|1, 0, n-1\rangle$, $|0, 1, n-1\rangle$, $|1, 1, n-2\rangle$, $|0, n\rangle$ with the notation $|n_{X\uparrow}, n_{X\downarrow}, n_P\rangle$, where $n_{X\alpha}$ (n_P) is the exciton (photon) number, takes the form of

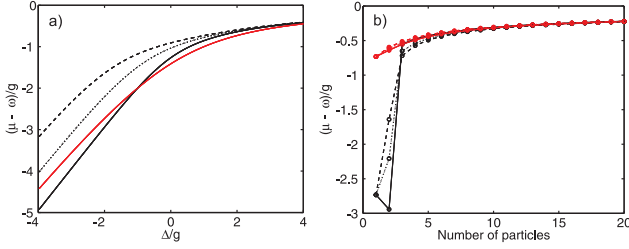


Fig. 9.7 a): Chemical potential as a function of exciton-photon detuning Δ for $t_X = 0$ and $t_P = 0$, and for different number of particles $n = 1$ (red [gray]) and $n = 2$ (black). b): Chemical potential as a function of the number of particles for different detunings $\Delta = -2g$ (black) and $\Delta = 2g$ (red [gray]), and for $t_X = 0$ and $t_P = 0$. Various on-site energies $W = -g$ (solid), $W = 0$ (dotted), and $W = g$ (dashed) are taken into account.

$$H_{IN} = \begin{pmatrix} \Delta & 0 & \sqrt{n-1}g & \sqrt{ng} \\ 0 & \Delta & \sqrt{n-1}g & \sqrt{ng} \\ \sqrt{n-1}g & \sqrt{n-1}g & 2\Delta + W & 0 \\ \sqrt{ng} & \sqrt{ng} & 0 & 0 \end{pmatrix}, \quad (9.47)$$

with the ground state energy $E_{IN}(n, \Delta)$. Its knowledge, as a function of the number of particles or detuning, enables to calculate the chemical potential from its definition as an energy needed to add a new particle into the system

$$\mu(n, \Delta) = E_{IN}(n+1, \Delta) - E_{IN}(n, \Delta). \quad (9.48)$$

This is the starting point of our analysis of phase diagrams in the next section and it is shown in Fig. 9.7.

Before we focus on the numerical results, we note that in order to explore experimentally the phase diagrams shown in the next section, it is necessary that exciton and photon transfer energies t_X and t_P vary over several orders of magnitude. In the exciton case, as already mentioned above, this can be achieved e.g. by modifying the Förster transfer [39] or by applying external fields [48]. Varying photon transfer energies is however more difficult since assuming that photon hopping limits the cavity quality factor Q then it holds $t_P = \omega_C/Q$ [41]. Consequently, a post-creation precise control of the quality factor of each cavity is required.

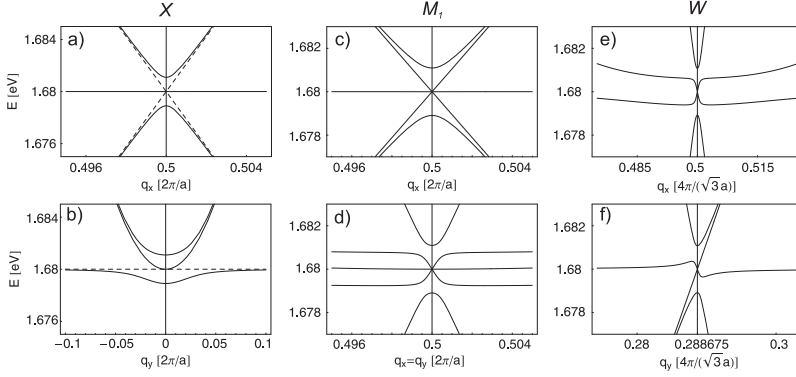


Fig. 9.8 The energy dispersion of the Bragg polaritons at selected high symmetry points. X -polaritons along $\bar{\Gamma}X$ (a) and along $X\bar{M}$ (b), M_1 -polaritons along $\bar{\Gamma}M$ (c) and $q_x=q_y$ axis (d), and W -polaritons along q_x -axis (e) and q_y -axis (f). The dashed lines in (a) and (b) represent the uncoupled exciton and photon modes.

9.3 Results and discussion

9.3.1 Neutral quantum dot lattice

9.3.1.1 Ideal lattices

We start by presenting the numerical results for Bragg Polaritons at three high symmetry points of the square and hexagonal lattice (see Fig. 9.3) in the ideal case (no disorder). The numerical calculations include all resonant and off resonant terms and parameters can be found in [5]. We note that the cavity length, dot size, and lattice constant can be optimized in order to maximize the light-matter coupling for Bragg polaritons at a given high symmetry point [5].

In Fig. 9.8 we show the energy dispersion of the Bragg polariton modes for X -polaritons, M_1 -polaritons, and W -polaritons. The different behavior of the upper polariton (UP), central polariton (CP), and lower polariton (LP) modes at different points shows how richer Bragg polaritons are compared to the QW polaritons. Note that the total number of branches crossing or anti-crossing at the different symmetry points is always given by $n + 1$. At the X -point, we have $n = 2$ and consequently there are three polariton modes. In the q_y -direction, the upper and lower X -polaritons are similar to the QW polariton (see Fig. 9.8b). However, there is one additional central mode degenerate with the photon mode along the zone boundary in the q_y -direction. By contrast, the q_x dependence along $\bar{\Gamma}X$ shows the existence of (i) a lower branch with a negative effective mass in one direction, (ii) an upper branch with positive effective masses but high anisotropy, and (iii) a flat branch with a large mass (consequence of the infinite exciton mass considered in our model) with energy exactly between the lower and upper branch. Let us now look at the M_1 -polaritons, which have five branches. In the $q_x = q_y$ direction we can see

nearly flat photonic branches originating from photon modes with $\mathbf{Q} = (2\pi/a, 0)$ and $\mathbf{Q} = (0, 2\pi/a)$, while the polariton mixing occurs mainly with photon modes having $\mathbf{Q} = (0, 0)$ and $\mathbf{Q} = (2\pi/a, 2\pi/a)$. Notice that there is not a saddle point at the M -point. Polariton mode with isotropic negative mass (lower branch) can give rise to an effective negative index of refraction [49]. In the W -point case for the hexagonal lattice there are four branches and the properties are similar to the M -point of the square lattice.

In the case of QW polaritons, the excitonic component is equally shared between lower and upper polaritons at the anticrossing. This is also true for the excitonic component in the X -point as shown in Fig. 9.9 (a and c). At the anti-crossing, the UP and LP branches consist of *half exciton* and consequently, the excitonic component is completely absent in the CP branch. Nevertheless, it increases rapidly away from the anticrossing region. This exciton-photon character swap is a characteristic of the Bragg polaritons since in all the cases investigated we have found analytically and numerically that the excitonic component is always equally shared between the lower and upper polaritons at the anticrossing, the rest being purely photonic.

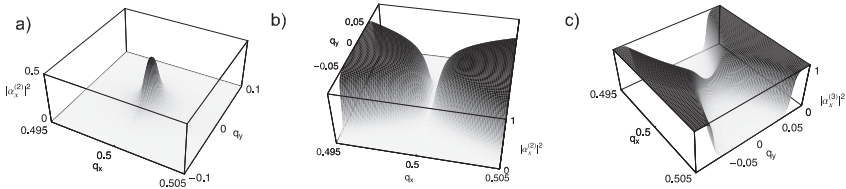


Fig. 9.9 The exciton component of the lower (a), central (flat) (b), and upper (c) Bragg polariton at the X -point.

Inhomogeneities in the quantum dot system are expected to affect the results presented so far. If the light-matter interaction is not very strong, Bragg polaritons are sensitive to three kinds of inhomogeneities: energy and oscillator strength fluctuations, and fluctuations in the position due to the deviations from an ideal lattice. In the following we will discuss the role of these three kinds of disorder.

9.3.1.2 Disorder effects

Here we present the absorption spectra calculated numerically for various strengths of disorder and averaged over many disorder realizations. We consider the case of an excitation at the M point (finite excitation angle corresponding to $q_M = (\pi/a, \pi/a)$) and at the Γ point (normal excitation). The exciton energy is tuned into resonance with the corresponding cavity modes. Therefore, the calculations for Γ and M point refer to different structures with lattice constants a and cavity length L_C adjusted to obtain a strong coupling at the two points as can be found in [5]. The full Hamiltonian corresponding to a lattice with 31^2 sites and photon modes in an energy window $[0.9\omega_X, 1.1\omega_X]$ is diagonalized. In order to make the results independent of the

system size we keep the coupling constant $\tilde{g}_q$ fixed and we use periodic boundary conditions.

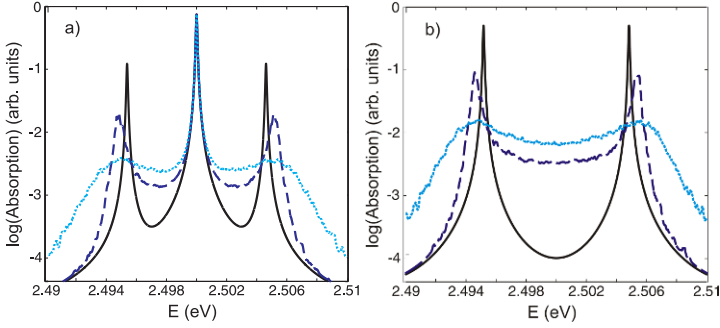


Fig. 9.10 Polariton absorption spectrum in a logarithmic scale for energy disorder with variance 0 meV (black [solid]), 2 meV (blue [dashed]), and 4 meV (cyan [densely dotted]) for polaritons at two high symmetry points of the first BZ: M (a) and Γ (b). A homogeneous Lorentzian broadening of $\gamma=50\mu\text{eV}$ was considered. Material parameters correspond to ZnSe/CdSe systems, and can be found in [5].

Let us start with the effect of the energy disorder in the absorption spectra as shown in Fig. 9.10. The disorder-free case (black line) shows lower (LP) and upper (UP) polariton peaks with half-exciton and half-photon character. There is an additional central peak in the M -point case (a). This peak arises from the mixing of photonic modes that differ by reciprocal lattice vectors. For symmetry reasons one of those mixed states is decoupled from the quantum dots at the M -point and we call this peak "photonic". The disorder shifts the LP (UP) energy: According to perturbation theory the shift of the LP (UP) is zero to first order and is negative (positive) to second order. Therefore, we expect an *increase* of the polariton splitting. Weak disorder can increase the robustness and visibility of the polariton splitting with respect to the disorder-free case. The fact that a weak exciton disorder increases the observability of a coherent effect may be seen as counter-intuitive, but the same effect has also been found in similar systems, e.g. in the exciton Aharonov-Bohm effect [50, 45]. Notice that for both Γ and M polaritons, a central very broad excitonic peak appears due to disorder. In the M -point case, however, a central peak insensitive to the disorder is superimposed on the broad central excitonic background. As the width of the inhomogeneous energy distribution approaches the value of the lower to upper polariton splitting, the three peaks merge. In the strong disorder limit the spectrum consists of a central narrow (photon-like) peak and a broad excitonic background.

Recent systems with arrays of quantum dots with inhomogeneous broadening of the order of 1 meV have been reported in [51]. However fluctuations in the oscillator strength may still be present. In order to investigate this effect, we consider a Gaussian distribution of the exciton oscillator strength in the coupling constants g_{jq} . The absorption spectra for different oscillator strength disorder are shown in Fig. 9.11.

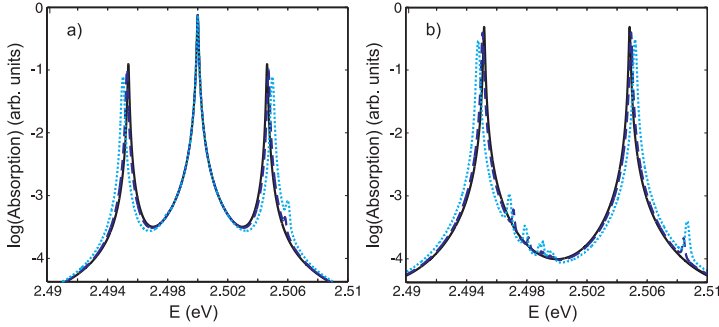
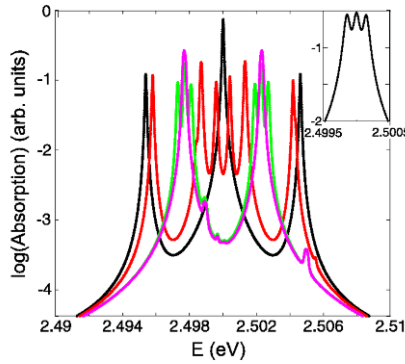


Fig. 9.11 Polariton absorption spectrum in a logarithmic scale for oscillator strength disorder with variance 0 % (black [solid]), 20 % (blue [dashed]), and 40% meV (cyan [densely dotted]). Polaritons at two high symmetry points of the first BZ are shown: M (a) and Γ (b). Material parameters as in Fig. 9.10.

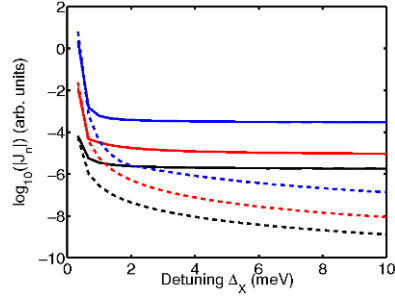
The behavior of the LP and UP peaks for both Γ and M -point is similar to the case of energy disorder. In particular, we observe a decrease (increase) of LP (UP) energy. We also remark that even for very large fluctuations of the oscillator strength both the broadening and energy shift remain smaller with respect to the inhomogeneous energy disorder. The additional peaks seen between and above the two main peaks are caused by polariton multiple scattering, i.e. scattering involving polariton states with different q . Only few peaks are visible because of the finite size of the system in the numerical simulation. The asymmetry reflects the polariton band dispersion. Note that in the case of quantum wells multiple scattering effects have been shown to be negligible [52]. Here we have an example of a disorder for which the full q dependence of the polaritonic states is important.

Fig. 9.12 Polariton absorption spectrum in a logarithmic scale for position disorder with variance from 0 (black, no side peaks) to 10% (red), 30% (green), and 50% (magenta, strongest side peaks) of the lattice constant. Polariton modes at the M point of the first BZ are considered. Inset: Detail of the central polariton peak for 1% disorder. Material parameters as in Fig. 9.10.



Finally, we have also investigated absorption spectra in the presence of positional disorder, i.e. deviations in the exciton localization sites from the ideal lattice, as shown in Fig. 9.12. Clearly, this disorder plays a role only for M -point polaritons, since the Γ -point does not probe the lattice symmetry. In contrast to the os-

Fig. 9.13 Logarithmic plot of $J_R^{(n)}$ (solid) and $J_O^{(n)}$ (dashed) as a function of the exciton detuning Δ_X in a 3×3 array of charged QDs with $g = 100 \mu\text{eV}$, $J_S = 0.43 \text{ meV}$, $V_{LP} = 10 \mu\text{eV}$, and $\delta = 50 \mu\text{eV}$. From bottom to top: $n = 2$ (black), $n = 4$ (red), and $n = 6$ (blue).



cillator strength disorder and to the energy disorder, positional disorder leads to a blueshift (redshift) of the lower (upper) polariton peak, which implies a reduction of the polariton splitting. The behavior of the central polariton (CP) peak is more complex. First, it splits into three lines for weak disorder (see inset of Fig. 9.12). Then the remaining central peak splits again into a doublet for stronger disorder (red curve in Fig. 9.12). The first splitting can be understood taking into account the CP's three-fold degeneracy [5] at q_M which is removed in the presence of disorder. Consequently, one mode is blue-shifted, one mode is red-shifted, and the third one remains unchanged. By further increasing the disorder strength, the state with q_M mixes with neighboring q and a doublet appears. If the positional disorder is of the same order as the lattice constant, i.e. for $\sigma_R \sim 0.5a$, the lattice becomes equivalent to random distribution of sites. In this limit, the random phase e^{iqR_j} in the coupling constants can be averaged in the Hamiltonian, which leads to a position independent value $\langle g e^{iqR_j} \rangle = g$. This results in a two-peak spectrum that qualitatively looks like the one of the Γ point resonant system with an upper and lower polariton as clearly seen for the strongest disorder in Fig. 9.12 (magenta). Such a spectrum with only two peaks is also obtained in a quantum well-microcavity for arbitrary values of the angle and detuning.

9.3.2 Charged quantum dot lattice

Here we present some numerical results on the multi-spin coupling and gate errors in a lattice of charged quantum dots. Most of the parameters used in the calculations are given in the respective captions of the figures. The maximum exciton detuning is set to $\Delta_X = 10 \text{ meV}$, which is about the upper limit for a Stark shift that can be obtained in current experiments. For simplicity, we assume the same exciton and photon decay rates of $\Gamma = \kappa = 2 \mu\text{eV}$, which requires a very high Q ($\sim 10^6$) cavity, but is of the right order of magnitude for self-assembled QDs. In the numerical calculation we consider a finite system with 9 dots. First, the dependence of *real* parts of different multi-spin terms on the detuning is shown in Fig. 9.13 where we separate the terms that involve the two dots nearly resonant with the

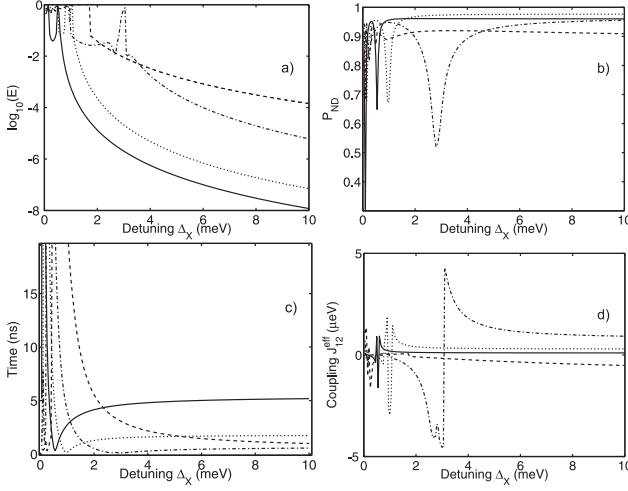


Fig. 9.14 Logarithmic plot of the error $\mathcal{E}$ (a), the probability of no state decay P_{ND} during the computation (b), the time of the computation t_C (c), and the effective coupling constant $\tilde{J}_{12}^{(2)}$ (d) as a function of the exciton detuning Δ_X . The phase gate between two dots in an array of nine charged QDs, $g = 100 \mu\text{eV}$, $\delta = 50 \mu\text{eV}$, $V_{LP} = 10 \mu\text{eV}$, $J_S = 0.86 \text{ meV}$ (dashed), $J_S = 0.43 \text{ meV}$ (dashed-dotted), $J_S = 0.22 \text{ meV}$ (dotted), and $J_S = 0.11 \text{ meV}$ (solid).

cavity from the other terms related to the off-resonant dots. We plot $J_{12} + J_{21}$ (solid black) and $\sum_{ij \notin \{1,2\}} |J_{ij}|$ (black dashed) for $n = 2$ spin terms. For multi-spin terms ($n = 4, n = 6$) the contributions that renormalize the effective coupling between the two resonant dots 1 and 2 ($J_R^{(n)}$) are separated from contributions that involve only the off-resonant dots strongly detuned from the cavity ($J_O^{(n)}$). For instance, for $n = 4$ the resonant (off-resonant) terms are defined as $J_R^{(4)} = \sum_P |J_{P(1122)}^{(4)}| + \sum_{iP} |J_{P(12ii)}^{(4)}|$ ($J_O^{(4)} = \sum_{ijkl} |J_{ijkl}^{(4)}| - |J_R^{(4)}|$). This definition enables us to better estimate the contribution of the off-resonant terms. In fact, even if the magnitude of the individual terms $J_{i_1 \dots i_n}^{(n)}$ is small there is an enhancement due to the large number of n -dot combinations ($\sim \binom{n}{N_D}$). Note that although the magnitude of the resonant term increases with n since $J_R^{(n)} \sim \frac{J_S^n V_{LP}^2}{\delta^{(n+1)}}$ and the ratio $J_S \gg \delta$ dominates over $\delta \gg V_{LP}$, there is only a weak dependence on the exciton detuning for the resonant terms and a strong decrease for the off-resonant terms ($J^{(n)} \sim \frac{J_S^n V_{LP}^2}{\Delta_X^{(n-1)}}$) as expected from the form of the coupling in Eqs. (9.35) and (9.36).

Second, we show in Figs. 9.14 the calculated error (see Sec. 9.2.2.2) $\mathcal{E} = 1 - \mathcal{F}$, the probability P_{ND} of no state decay during a gate with time t_C (no quantum jump), and the effective spin-spin coupling $\tilde{J}_{12}^{(2)}$ as a function of the exciton detuning for different spin energy J_S . We notice that the error tend to decrease with increasing exciton detuning Δ_X since at larger detunings only the two selected dots $\{1, 2\}$ re-

main in resonance with the cavity and the multi-spin coupling with the other dots is suppressed.

Another clear trend at large Δ_X is the decrease of the computation time with increasing the spin energy (Fig. 9.14c). We note that for sufficiently strong exciton detuning, the operation times are $t_C < 5$ ns. Thus, they are shorter than the spin decoherence time T_2 , which is of orders of at least μ s. Moreover, it turns out that the optimal way to decrease the computation time t_C for large Δ_X within the assumptions $\gamma \ll \delta \ll J_S$ and $V_{LP} \ll \delta'$ (detuning with respect to the polariton-spin levels) is to increase the spin energy J_S . The behavior of the probability P_{ND} is more complicated since it is affected by the strength of the spin interaction $J_{12}^{(2)}$, by the time of computation t_C , and by the distance between the laser energy ω_L (or detuning δ) and the energy of the trions $\omega_{X,1} \pm \frac{J_S}{2}$. In general, the probability of no-decay P_{ND} should increase by decreasing the laser detuning δ as follows from Eq. (9.38). On the other hand, since the condition $\delta < J_S$ has to be fulfilled, the probability of the no-decay increases by increasing J_S (up to the certain value of $\frac{\delta}{J_S}$). Now we turn our attention on the resonant features in the effective coupling $\tilde{J}_{12}^{(2)}$ as shown in Fig. 9.14d. A similar resonant-like behavior has been already predicted in the case of the spins localized by impurity centers in a semiconductor host [30]. Here, the features are caused by spin multiplets, which for small values of the exciton detuning Δ_X are mixed with the spins of the resonant dots through $\tilde{J}_{12ij}^{(4)}$ and higher order terms. Since each spin multiplet has its own decay rate γ and characteristic dependence on the spin energy J_S , it can happen that the laser energy is close to one of the spin multiplet energies. When this happens, it (i) increases the effective spin-spin coupling, (ii) increases the decay rate, (iii) decreases the fidelity, and (iv) decreases the time of computation.

9.3.3 Neutral quantum dots in lattice of cavities

Here we present numerical results on the array of cavities containing single dots. We discuss the phase diagrams (order and coherence parameters) and the numbers of excitons or photons as functions of chemical potential, exciton and photon hopping energy, t_X and t_P . We have taken into account up to 20 particles (either excitons or photons) and minimized the system energy with respect to the order parameter ψ and the coherence parameter χ .

Let us start looking at the dependence of chemical potential μ on detuning Δ and number of particles n , which is shown in Fig. 9.7. We can notice that for negative detunings and biexciton case $W < 0$, it holds for the chemical potential, $\mu(2, \Delta) < \mu(1, \Delta)$, and the system changes its state directly from $n = 0$ to $n = 2$, without passing through one particle state. In the case of repulsion (no biexciton), there are distinguishable transitions from zero to one and from one to two particles, which, however, get closer with positive Δ as the particles tend to be more photon-like and therefore, less sensitive to the exciton potential W . The transitions for more

particles $n > 2$ are smooth as only the number of photons is increased (having always two excitons). In the limiting case of a very large number of particles $n \rightarrow \infty$, the chemical potential approaches zero $\mu(\infty, \Delta) \rightarrow 0$, and consequently the system approaches the noninteracting (photon) Bose gas.

Before we start to discuss the meanfield results, we note that they are valid only for $\mu < \omega_C$, where ω_C the cavity photon mode energy, since for $\mu \rightarrow \omega_C$ the method does not converge. However, in the case of $\mu \rightarrow \omega_C$ the system is superfluid. We further note that the exciton spontaneous emission rate γ_X or the photon leakage rate out of the nanocavity κ lead to an effective broadening of eigenstates. In order to observe Mott lobes, the following condition has to be fulfilled $\mu(n+1, \Delta) - \mu(n, \Delta) > \Gamma$ with Γ being the ground state broadening. This means that effectively only the lowest Mott lobes are easy to observe.

9.3.3.1 Exciton phase diagrams

We begin with exciton phase diagrams. We note that without light-matter interaction, $g = 0$, and with exciton-exciton repulsion, $W > 0$, the meanfield results of the Hubbard model [26], i.e. three Mott lobes instead of two (as we will see), are retrieved.

First, we take a very small photon hopping (coupling) parameter $t_P \ll g$ and focus on phase diagrams for the order parameter ψ for negative detuning Δ as a function of chemical potential μ and normalized exciton transfer energy t_X/g , which are depicted in Fig. 9.15 a). Due to our choice of photon hopping, the system is effectively only excitonic for most values of the chemical potential. There are clearly visible Mott lobes with $\psi = 0$ for small t_X and superfluid phase with $\psi \neq 0$ for large t_X . The boundaries of Mott lobes for a small transfer t_X can be identified from the dependence of the chemical potential on the number of particles in Fig. 9.7. In the biexciton case $W < 0$, there are only two lobes, either without any particle or with a biexciton as seen in Fig. 9.15 a). In the case of exciton repulsion, there would be three lobes with zero, one, and two particles, respectively. This would resemble either the results of the pure Hubbard model [53] or the case of coupled cavities although in that case there are many modes since there is no limitations on the number of photons [20]. Furthermore, the number of photons starts to increase only as the chemical potential approaches zero in correspondence with results seen in Fig. 9.7. Moreover, for these values of chemical potential the photon superfluidity appears. This means that order and coherence parameters are *correlated* in a nontrivial way. In the one-component system, only the absolute value of the order or coherence parameter is fixed and usually plus sign is chosen due to the intuitive physical interpretation $\sqrt{n_{j\alpha}} = \langle C_{j\alpha} \rangle = \langle C_{j\alpha}^\dagger \rangle$. Here, this unambiguity is partially broken, since only one of the parameters can be positive, i.e. a new additional condition for energy minimum appears

$$\psi \cdot \chi < 0. \quad (9.49)$$

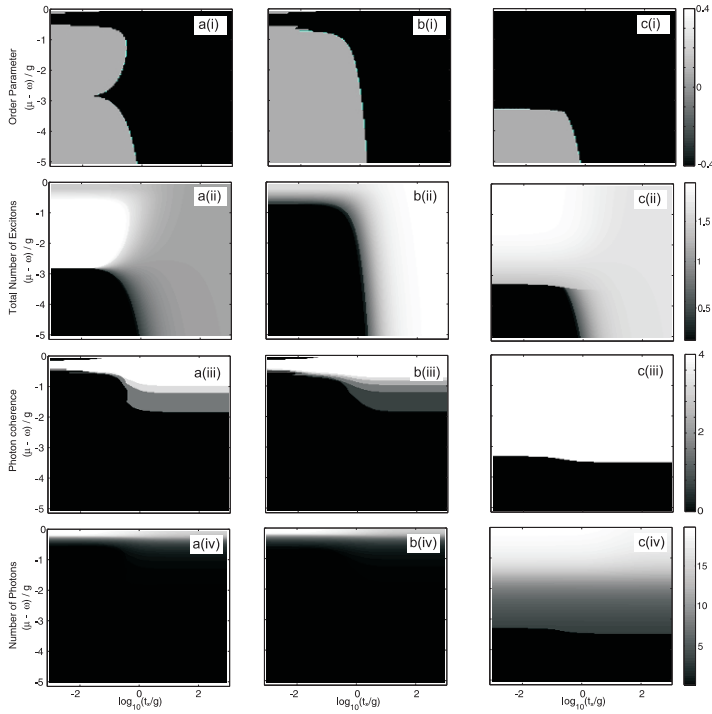


Fig. 9.15 Order parameter ψ (i), number of excitons n_X (ii), photon coherence parameter χ (iii), and number of photons n_P (iv) as a function of the exciton tunnelling (coupling) parameter t_X and chemical potential μ for negative detuning $\Delta = -2g$ [a] and c)] and $\Delta = 2g$ [b)], for on-site exciton energy $W = -g$, and for photon hoppings: $t_P = 10^{-3}g$ [a] and b)] and $t_P = g$ [c)]. In the case of a nonconstant behavior, the insulating (superfluid) phases, i.e. $\psi = 0$ ($\psi \neq 0$) or $\chi = 0$ ($\chi \neq 0$), are found on the left-hand (right-hand) side.

One can thus choose $\psi > 0$, which implies $\chi < 0$ and the naive interpretation of the coherence parameter as $\chi = \sqrt{n_j^P}$, n_j^P being the number of photons, is lost.

Additionally, the case of positive detuning is shown in Fig. 9.15b). In this case results for different exciton potentials W are similar since there are only two exciton phases: (i) insulating without any particle and (ii) superfluid. As in the previous case, depending on whether there are superfluid photons or not in the system, the signs of the order and coherence parameters are correlated. Moreover, the number of particles increases with μ or when the total exciton energy becomes comparable to the lower photon energy due to increased transfer t_X .

Second, we proceed further by increasing the photon hopping to the level of light-matter coupling $t_P = g$ (medium value of hopping). Corresponding results are plotted in Fig. 9.15c). Unlike in the first case, profound differences to the scenario which is usually seen for fermionic or bosonic Hubbard models are found. The fact that the system consists of two kinds of particles, fermionic excitons and bosonic photons, manifests itself strongly since there are clearly two ways of changing the

ground state nature: (i) Insulator-superfluid transition as the exciton order parameter changes from $\psi = 0$ to $\psi \neq 0$ with increasing exciton hopping t_X . (ii) Transition from the purely excitonic ground state (caused by negative detuning) with $\psi = 0$ or $\psi \neq 0$ for small chemical potential μ to the mixed exciton-photon state with sign correlation for a larger chemical potential μ . Moreover, photons (if present) are always found in the superfluid phase since the value of photon hopping, $t_P = g$, is sufficient to overcome the on-site effective photon repulsion due to the coupling to excitons [20]. The increase of photon number slightly depends on exciton hopping t_X , the larger it is the more preferable excitons are. Increasing the photon hopping even further, $t_P \gg g$, leads the system to the purely photonic and superfluid phase as expected (not shown). This case is interesting from the point of view of the polariton (exciton-photon) Bose-Einstein condensation (BEC) in microcavities whose investigation has gained a considerable interest in recent years both theoretically (see [54] and references therein) and experimentally [2, 3, 55, 56, 57]. Nevertheless, in the theoretical investigation of polariton condensation [58, 59, 60] it is always assumed that the photon field is superfluid and the exciton part is further investigated. Even though the assumption of photon superfluidity is well justified in planar microcavities where $t_P \gg t_X$ [21] (as in the current case), it leads to the conclusion that polariton condensation is more similar (possibly identical) to polariton laser coherence [61, 62], i.e. $\chi \neq 0$, than to (nonequilibrium) Bose-Einstein condensate of the matter [63]. In other words, if photon coherence (superfluidity) disappeared there would be no condensation. This critical point of view has been recently formulated by Butov in [64]. As our results show, the exciton superfluidity would remain for a sufficiently small exciton mass, approximately holding $g \sim t_X \sim \frac{\hbar^2}{2m_X d^2}$, where d is the interdot distance and m_X the effective exciton mass. For reasonable values of $g = 0.1 \text{ meV}$ and $d = 100 \text{ nm}$, one obtains that a very light exciton mass of $m_X = 0.03 m_0$ (m_0 being the bare electron mass) is needed. Such a light mass is difficult to find in typical semiconductor nanostructures. We may also ask if condensation of any composite system (e.g. exciton-photon) is possible at all. Our results show that genuine condensation, where coherence is present for both particles simultaneously, is possible only if the hopping energy of both constituents does not differ very much (within one order of magnitude). From this perspective, it is also understandable that one can observe exciton Bose-Einstein condensation since, in the language of the present model, hopping energies of electron and hole are within the same order of magnitude. We note that the investigation of exciton BEC in coupled quantum wells has gained a lot of experimental interest recently [65, 66].

9.3.3.2 Photon phase diagrams

After studying phase transitions from the exciton perspective we now turn our attention towards the photon perspective. As already mentioned in the introduction quantum phase transitions of light have become a very active field of research only very recently. Although we are going to plot the results in the same way as for QPT

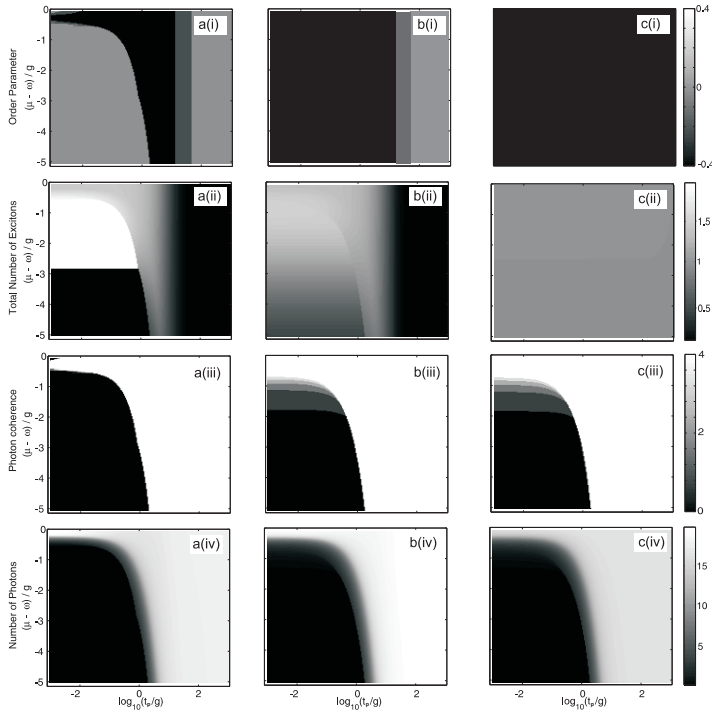


Fig. 9.16 Order parameter ψ (i), number of excitons n_X (ii), photon coherence parameter χ (iii), and number of photons n_P (iv) as a function of photon tunnelling (coupling) parameter t_P and chemical potential μ for negative detuning $\Delta = -2g$, for on-site exciton energy $W = -g$, and for exciton hoppings: $t_X = 10^{-3}g$ [a)], $t_X = g$ [b)], and $t_X \gg g$ [c)]. In the case of a nonconstant behavior, the insulating (superfluid) phases, i.e. $\psi = 0$ ($\psi \neq 0$) or $\chi = 0$ ($\chi \neq 0$), are found on the left-hand (right-hand) side.

of light it is important to keep in mind that we are dealing with a coupled exciton-photon system.

As in the previous section we begin with the almost one particle system, i.e. $t_X \ll g$, with the results depicted in Fig. 9.16a. Indeed, these results resemble very much those for the system studied for quantum phase transitions of light. Especially, the plot of photon coherence in Fig. 9.16a shows many common features with Fig. 4c in [20], also calculated for negative detuning. A clear insulator-superfluid transition for photons can be observed. However, there are two main differences to previous studies: (i) In the absence of photons, the system behaves as an exciton system, i.e. due to our choice of the exciton-exciton interaction, as mentioned above, there is an immediate transition from zero to two excitons (biexciton) with increasing chemical potential μ . (ii) As we have seen before, the order and the coherence parameters are correlated and thus, with superfluid photons found in the system, the exciton is also in the superfluid phase. Nevertheless, with increasing photon hopping t_P the exciton component disappears.

Furthermore, results for an increased exciton hopping energy, $t_X = g$, are shown in Fig. 9.16b. An insulator-superfluid transition of light can again be nicely seen, which is only very slightly modified by the increased exciton hopping with respect to the results plotted in Fig. 9.16a. The increased exciton hopping t_X leads to the fact that for small photon hopping t_P there is only superfluid exciton found in the system ($\Psi \neq 0$) and as soon as the superfluid photon appears the sign correlation is established. The exciton then gradually disappears as the photon hopping t_P , and consequently the photon number n_P , is increased.

Last but not least, the exciton hopping energy is increased even further and results are shown in Fig. 9.16c). The scenario from the previous case is repeated. However, the superfluid exciton is found in the system for all values of the hopping t_P since a sufficiently strong value, which is needed to dominate and force the system to be purely photonic and superfluid, is not reached. Finally, we mention that the (energy, oscillator strength, positional) disorder [22] would introduce a third phase similar to the Bose glass in the case of the Bose-Hubbard model [53].

9.4 Conclusions

We have investigated excitons and trions in a two-dimensional quantum dot lattice embedded in a planar optical cavity. The strong exciton (trion)-photon coupling has been described in terms of polariton quasiparticles with extremely small mass, which makes them the lightest exciton-like quasiparticles in solids. The effect of disorder has been also discussed. Furthermore, one electron has been added into each dot. Such a “spin lattice” can be used for quantum information processing and we have shown that by using exciton detuning a conditional phase shift gate with a high fidelity can be obtained. Finally, we have investigated exciton-photon quantum phase transitions in a planar lattice of one-mode cavities containing one quantum dot. We have demonstrated that by controlling exciton- and photon-hopping energies a very rich scenario of coupled fermionic-bosonic quantum phase transitions appears. We have discussed its relationship to the interpretation of the Bose-Einstein condensation of polaritons.

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Part IV
**Coherent light-matter states in
semiconductor microcavities**

Chapter 10

Quantum optics with interacting polaritons

Stefano Portolan and Salvatore Savasta

Abstract The excitonic polariton concept was introduced already in 1958 by J. J. Hopfield. Although its description was based on a full quantum theory including light quantization, the investigations of the optical properties of excitons developed mainly independently of quantum optics. In this chapter we shall review exciton polariton quantum optical effects by means of some recent works and results that have appeared in the literature in both bulk semiconductors and in cavity embedded quantum wells. The first manifestation of excitonic quantum-optical coherent dynamics was observed experimentally 20 years later, in 1978, exploiting resonant hyper-parametric scattering. On the other hand, the possibility of generating entangled photon pairs by means of this resonant process was theoretically pointed out only lately in 1999, whereas the experimental evidence for the generation of ultraviolet polarization-entangled photon pairs by means of biexciton resonant parametric emission in a single crystal of semiconductor CuCl was reported only in 2004. The demonstrations of parametric amplification and parametric emission in semiconductor microcavities, together with the possibility of ultrafast optical manipulation and ease of integration of these micro-devices, have increased the interest in possible realization of nonclassical cavity-polariton states. In 2005 an experiment that probes polariton quantum correlations by exploiting quantum complementarity was proposed and realized. These results unequivocally proved that quantum optical effects at single photon level, arising from the interaction of light with electronic excitations of semiconductors and semiconductor nanostructures, were possible within these solid state systems, despite being far from isolated systems. The theoretical predic-

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tions that we review are based on a microscopic quantum theory of the nonlinear optical response of interacting electron systems relying on the dynamics controlled truncation scheme extended to include light quantization. In addition, the environment decoherence is taken into account microscopically by Markov noise sources within a quantum Langevin framework.

10.1 Introduction

The field of quantum optics has witnessed relevant developments and has raised a lot of interest in the last decades. Since the pioneering studies on the coherence properties of radiation to modern quantum optical experiments — addressing fundamental issues of quantum mechanics such as Bell's theorem, quantum nondemolition measurements and quantum complementarity — the subtle properties of light have been deeply debated topics. Recent proposals [1] and realizations [2, 3, 4, 5, 6] of many-particle entangled quantum states require better understanding of the domain of validity of quantum behaviour. Moreover, atom-cavity systems have been used to investigate quantum dynamical processes for open quantum systems in a regime of strong coupling and to explore quantum behaviours that have no classical counterparts [7]. On the other hand, since the early seventies [8] researchers have been exploring possible realization of semiconductor-based heterostructures, devised according to the principles of quantum mechanics. The development of sophisticated growth techniques started a revolution in semiconductor physics, determined by the possibility of confining electrons in practical structures. In addition, the increasing ability to control fabrication processes has enabled the manipulation of the interaction between light and semiconductors by engineering not only the electronic wave functions but also the photon states. The radiation rate of an excited atom can be controlled by changing the distribution of electromagnetic modes near the atom using a cavity [9]. In semiconductor microcavities (SMCs), the interaction between light and excitons can be engineered in exciting and subtle ways. When one or more QWs are embedded in an optical semiconductor micrometric spacer embedded between two distributed Bragg reflectors the coupling between the light (cavity modes) and the exciton can be tailored to be more effective than the relaxation mechanisms. This is the case of what is known as strong-coupling regime for cavity exciton-polaritons. The modification of the spontaneous emission in a cavity and the strong-coupling regime are cavity quantum electrodynamics effects already well known in quantum optics. This is some of the evidence for the strong similarities existing between semiconductor microcavities and atomic cavity-systems.

However, the exciton-cavity system originating from this heterostructure has some fundamental differences with respect to the simpler two-level atom-single mode cavity model, greatly exploited in quantum optics, even though the former displays a very similar coherent linear dynamics despite of the complexity of the electronic semiconductor states. Indeed when a weak light beam of a given wave vector excites the electronic system, only one-exciton states interact and, owing to

customary conservation rules, this exciton has the same wave vector of the impinging beam. On the contrary, in contrast to the two-levels atomic systems where the source of nonlinearities comes from saturation, the nonlinear dynamics in semiconductor heterostructures, mainly due to Coulomb correlation between electrons, does not maintain this simple picture. Laser spectroscopy in semiconductors and in semiconductor quantum structures, gives access to the physics of coherences, correlations and quantum kinetics involving charge, spin and lattice degrees of freedom. It has been greatly exploited because an excitation with ultrashort optical pulses in general results in creation of coherent superpositions of many-particle states. Therefore, semiconductor heterostructures excited by ultrafast laser pulses are ideally suited to serve as prototype systems where quantum-mechanical properties of many interacting particles, far away from equilibrium, and of light can be studied in a controlled fashion. Thus it constitutes a very promising powerful tool for the study of correlation and an ideal arena for semiconductor cavity quantum electrodynamics (cavity QED) experiments as well as coherent control, manipulation, creation and measurement of non-classical states [10, 11, 12, 13]. The analysis of nonclassical correlations in semiconductors constitutes a challenging problem, where the physics of interacting electrons must be added to quantum optics and should include properly the effects of noise and dephasing induced by the electron-phonon interaction and the other environment channels [14]. The physical picture describing the dynamical evolution of optically excited electrons and holes can be described in simple terms as follows. First of all, the exciting laser field with frequency near the fundamental band gap creates coherent electron-hole (eh) pairs. Subsequently, the motion of the carrier population, dominated by Coulomb interaction, leads to an ultrafast electronic polarization (the source of the outgoing light that can be observed). The scattering processes due to other carriers, phonons, defects and the other degrees of freedom result in polarization decay and decoherence. Radiative recombination occurs on longer time scales thus the properties of the outgoing light manifest the underlying electronic dynamics. Ultrafast time resolved spectroscopy has been used as a versatile and powerful tool for investigating many-body effects in quantum structures. Moreover, ultrafast nonlinear optical response in semiconductor cavity-embedded QWs within a nonperturbative regime has been attracting growing interest for exploring fundamental open questions on light-matter interaction in many-body quantum systems, as well as for appealing future applications in optoelectronic and photonic devices.

10.2 Electronic excitation in semiconductor

A key role in the optical response of dielectric media at frequencies near the band gap is played by excitons. Indeed, when a direct band gap semiconductor is in the presence of an electromagnetic field of energy equal to the electronic band-gap, an electron can be promoted from the highest valence band to the lowest conduction band by absorption of a photon. When an electron is promoted to the conduction

band the vacancy left in the valence band changes the charge density and it can be described as a hole related to the excited electron, the prototype of a 2-body process. Indeed the correlation among electrons produces an attractive Coulomb interaction between the electron and the hole giving bound states with energy less than the energy gap of the semiconductor. These bound electron-hole states are known as excitons and modify substantially the optical properties of the semiconductor structure.

In semiconductors the many-particle ground state can be approximated by a single Slater-determinant (ensuring antisymmetry) corresponding to filled valence and empty conduction bands separated by a gap (we shall consider direct-gap semiconductors). It is advantageous to describe particles in conduction states by electron Fermi operators $\hat{c}_{\mathbf{k}}^\dagger$ ($\hat{c}_{\mathbf{k}}$) and to use hole Fermi operators $\hat{d}_{\mathbf{k}}^\dagger$ ($\hat{d}_{\mathbf{k}}$) to represent valence states, i.e. $\hat{c}_{\mathbf{k}}^\dagger$ creates ($\hat{c}_{\mathbf{k}}$ destroys) an electron in a conduction state, while $\hat{d}_{\mathbf{k}}^\dagger$ creates ($\hat{d}_{\mathbf{k}}$ destroys) a hole in a valence state. Here $\mathbf{k}$ are 3D wave-vectors in the first Brillouin zone, or when considering electrons in quasi-2D nanostructures (quantum wells) indicates 2D in-plane wave-vectors. More generally this label can be regarded as a collective quantum number specifying single-particle valence and conduction states in semiconductors or semiconductor quantum structures. This electron-hole picture has the advantage that the many-particle ground state serves as a vacuum state for electron and hole operators, resulting in particularly simple initial conditions for observables derived from these operators. In particular, when considering undoped semiconductors, the ground state is characterized by filled valence bands and empty conduction bands corresponding to a state with zero holes in valence bands and zero electrons in conduction bands. In the following we consider a zinc blende-like semiconductor band structure. The valence band is made from p -like ($l = 1$) orbital states which, after spin-orbit coupling, give rise to $j = 3/2$ and $j = 1/2$ decoupled states. In materials like bulk CuCl, the upper valence band is twofold degenerate ($j = 1/2$), while in materials like GaAs it is fourfold degenerate ($j = 3/2$). In GaAs-based quantum wells the valence subbands with $J = 3/2$ are energy split into two-fold degenerate heavy valence subbands with $m = \pm 3/2$ and lower energy light-hole subbands with $m = \pm 1/2$. The conduction band, arising from an s -like orbital state ($l=0$), gives rise to $j = 1/2$ twofold states. In the following we will refer to materials like CuCl or GaAs-based quantum wells and will consider for the sake of simplicity only twofold states from the upper valence band and lower conduction band. As a consequence electrons in a conduction band as well as holes have an additional spin-like degree of freedom as electrons in free space. When necessary both heavy and light hole valence bands or subbands can easily be included in the present semiconductor model. The standard model Hamiltonian near the semiconductor band-edge can thus be written as $\hat{H}_s = \hat{H}_0 + \hat{V}_{Coul}$ [10], which is composed of a free-particle part $\hat{H}_0$ and the Coulomb interaction $\hat{V}_{Coul}$. $\hat{H}_0$ is given by

$$\hat{H}_0 = \sum_{m,\mathbf{k}} \varepsilon_{m,\mathbf{k}}^{(e)} \hat{c}_{m,\mathbf{k}}^\dagger \hat{c}_{m,\mathbf{k}} + \sum_{m,\mathbf{k}} \varepsilon_{m,\mathbf{k}}^{(h)} \hat{d}_{m,\mathbf{k}}^\dagger \hat{d}_{m,\mathbf{k}}, \quad (10.1)$$

where $\hat{c}^\dagger$ and $\hat{d}^\dagger$ are the electron (e) and hole (h) creation operators and $\varepsilon^{(e)}$ and $\varepsilon^{(h)}$ the e and h energies. The Coulomb interaction is given by

$$\begin{aligned} \hat{V}_{Coul} = \sum_{\mathbf{q} \neq \mathbf{0}} V_{\mathbf{q}} \sum_{m, \mathbf{k}, m', \mathbf{k}'} & \left[\frac{1}{2} \hat{c}_{m, \mathbf{k}+\mathbf{q}}^\dagger \hat{c}_{m', \mathbf{k}'-\mathbf{q}}^\dagger \hat{c}_{m', \mathbf{k}'} \hat{c}_{m, \mathbf{k}} \right. \\ & \left. + \frac{1}{2} \hat{d}_{m, \mathbf{k}+\mathbf{q}}^\dagger \hat{d}_{m', \mathbf{k}'-\mathbf{q}}^\dagger \hat{d}_{m', \mathbf{k}'} \hat{d}_{m, \mathbf{k}} - \hat{c}_{m, \mathbf{k}+\mathbf{q}}^\dagger \hat{d}_{m', \mathbf{k}'-\mathbf{q}}^\dagger \hat{c}_{m', \mathbf{k}'} \hat{d}_{m, \mathbf{k}} \right], \quad (10.2) \end{aligned}$$

where $V_{\mathbf{q}}$ is the Fourier transform of the screened Coulomb interaction potential. The first two parts are the repulsive electron-electron and hole-hole interaction terms, while the third one describes the attractive eh interaction. The additional label m indicates the spin degrees of freedom. As we will see the spin degeneracy of the valence and conduction bands (subbands) play a relevant role in the nonlinear optical response of electronic excitations. Only eh pairs with total projection of angular momentum $\sigma = \pm 1$ are dipole active in optical interband transitions. In CuCl photons with circular polarizations $\sigma = +(-)$ excite e with $m_e = +1/2$ ($m_e = -1/2$) and h with $m_h = +1/2$ ($m_h = -1/2$). In GaAs QWs the situation is analogous: photons with circular polarizations $\sigma = -(+)$ excite e with $m_e = +1/2$ ($m_e = -1/2$) and h with $m_h = -3/2$ ($m_h = 3/2$). A many-body interacting state is usually very different from a product state, however a common way to express the former is by a superposition of uncorrelated product states. The physical picture that arises out of it expresses the *dressing* the interaction performs over a set of noninteracting particles. The general many-body Schrödinger equation for this Coulomb-correlated system is

$$\hat{H}_s | \Psi \rangle = (\hat{H}_0 + \hat{V}_{Coul}) | \Psi \rangle = E | \Psi \rangle, \quad (10.3)$$

with $| \Psi \rangle$ the global interacting many-body state of the whole Fock space and E its corresponding energy. The system Hamiltonian commutes with the total-number operators for electron and holes, i.e. $\hat{N}_e = \sum_{\mathbf{k}, m} \hat{c}_{\mathbf{k}, m}^\dagger \hat{c}_{\mathbf{k}, m}$ and $\hat{N}_h = \sum_{\mathbf{k}, m} \hat{d}_{\mathbf{k}, m}^\dagger \hat{d}_{\mathbf{k}, m}$. Therefore the state $| \Psi \rangle$ may be build up corresponding to a given number of electrons and of holes. Moreover, because we shall consider the case of intrinsic semiconductor materials where $N_e = N_h \doteq N$, the good quantum number for the Schrödinger equation (10.3) is the total number of electron-hole pairs N , explicitly

$$\hat{H}_s | N, \alpha \rangle = E_{N, \alpha} | N, \alpha \rangle, \quad (10.4)$$

where α is the whole set of proper quantum numbers needed to specify uniquely the many-body state. For any given number N of electron-hole pairs, the product-state set build up from the single-particle states $\{ | N, a \rangle \}$ — eigenstates of the noninteracting carrier Hamiltonian $\hat{H}_0$ — is a natural complete basis of the N -pairs subspace of the global Fock space:

$$\hat{H}_0 | N, a \rangle = \varepsilon_{N, a} | N, a \rangle, \quad (10.5)$$

where N identifies the N -pairs subspace and a is a compact form for all the single particle indexes, i.e. $a \equiv \mathbf{k}_{e1}, \mathbf{k}_{e2}, \dots, \mathbf{k}_{eN}; \mathbf{k}_{h1}, \mathbf{k}_{h2}, \dots, \mathbf{k}_{hN}$. Indeed

$$|N, a\rangle = \otimes_{n=1}^N \hat{a}_{c, \mathbf{k}_{en}}^\dagger \hat{a}_{h, \mathbf{k}_{hn}}^\dagger |0, 0\rangle \quad \text{and} \quad \varepsilon_{N, a} = \sum_{n=1}^N (\varepsilon_{\mathbf{k}_{en}} + \varepsilon_{\mathbf{k}_{hn}}). \quad (10.6)$$

Being a complete orthonormal basis for the N -pairs subspace we may expand the many-body state $|N, \alpha\rangle$ over it, it yields

$$|N, \alpha\rangle = \sum_a U_a^{N\alpha} |N, a\rangle. \quad (10.7)$$

It is only a matter of calculation to show that $U_a^{N\alpha}$ is nothing but the envelope function of the N -pair aggregate, solution of the corresponding Schrödinger equation in first quantization. Indeed the eigenvalue problem (10.4) is transformed into:

$$\sum_{a'} (\langle N, a | \hat{H}_s | N, a' \rangle - E_{N, \alpha} \delta_{a, a'}) U_{a'}^{N\alpha} = 0. \quad (10.8)$$

An important feature of $\hat{H}_s$ is that its matrix elements between states with a different number of eh pairs are zero. As a consequence it does not couple states with different numbers of eh pairs. The eigenstates $|N, \alpha, \mathbf{k}\rangle$ with energy $\omega_{N\alpha\mathbf{k}}$ of $\hat{H}_s$ can be labelled according to the number N of eh pairs and the total momentum $\mathbf{k}$. The state with $N = 0$ is the semiconductor ground state and corresponds to the full valence band. The $N = 1$ subspace is the exciton subspace. Optically active exciton states can be labelled with the additional quantum number $\alpha = (n, \sigma)$ where σ indicates the total spin projection (e. g. in CuCl $\sigma = +1$ indicates an e - h pair with $m_e = +1/2$ and $m_h = +1/2$; in GaAs QWs $\sigma = +1$ indicates an e - h pair with $m_e = -1/2$ and $m_h = 3/2$) and n spans all the exciton levels. Exciton eigenstates can be obtained by requiring that general one eh pair states be eigenstates of $\hat{H}_s$:

$$\hat{H}_s |N = 1, n\sigma, \mathbf{k}\rangle = \hbar\omega_{n\sigma}(\mathbf{k}) |N = 1, n\sigma, \mathbf{k}\rangle, \quad (10.9)$$

with $|N = 1, n\sigma, \mathbf{k}\rangle = \hat{B}_{n, \sigma, \mathbf{k}}^\dagger |N = 0\rangle$, $\hat{B}_{n, \sigma, \mathbf{k}}^\dagger$, being the exciton creation operator:

$$\hat{B}_{n, \sigma, \mathbf{k}}^\dagger = \sum_{\mathbf{k}'} \Phi_{n, \sigma, \mathbf{k}'}^* \hat{c}_{\sigma, \mathbf{k}' + \mathbf{k}/2}^\dagger \hat{d}_{\sigma, -\mathbf{k}' + \mathbf{k}/2}^\dagger. \quad (10.10)$$

In order to simplify a bit the notation, the spin notation in Eq. (10.10) has been changed by using the same label for the exciton spin quantum number and for the spin projections of the electron and hole states forming the exciton. The exciton envelope wave function $\Phi_{n, \sigma\mathbf{k}}$ can be obtained solving the secular equation obtained from Eq. (10.9). It describes the correlated electron-hole relative motion in $\mathbf{k}$ -space. As it results from H_s electrons and holes act as free particles with an effective mass determined by the curvature of the single-particle energies (bands or subbands) interacting via a (screened) Coulomb potential in analogy with an electron and a proton forming the hydrogen atom. Actually, it can be shown that the obtained $\Phi_{n, \sigma\mathbf{k}}$ is the Fourier transform of an hydrogen-like wave-function and the corresponding levels form an hydrogen-like spectrum with bound and unbound states:

$$\hbar\omega_{n\sigma}(\mathbf{k}) = E_n + \frac{\hbar^2}{2M}k^2, \quad (10.11)$$

where E_n describes the hydrogen-like spectrum originating from the electron-hole relative motion, while the dispersion term describes the centre of mass motion of the exciton M , being the sum of the electron and hole effective masses. The set of bound and unbound states with $N = 2$ *eh* pairs, determining the biexciton subspace, can be obtained solving the corresponding secular equation, or by using simpler interaction models. It is interesting to observe that the exciton subspace is spin degenerate. The exciton energy levels do not depend on the spin quantum number, hence the excitonic bands are doubly degenerate. The situation is more complex for the biexciton subspace. The structure of biexcitonic states and energies depends strongly on the spin of the involved carriers. In particular bound biexcitons that are the solid state analogue of the hydrogen molecules arise only when the two electrons and also the two holes have opposite spin. This is consequence of the Pauli exclusion principle. Bound biexcitons have energies that are usually some meV (or even more in large gap semiconductors) less than twice the energy of excitons. Biexcitonic states made by carriers with the same spin give rise only to a continuum starting at an energy that is twice that of the lowest energy exciton state. In order to analyze the dynamical evolution of the electronic polarization, it is useful to use the transition operators defined by

$$\hat{X}_{N,\alpha;N',\alpha'} = |N, \alpha\rangle \langle N', \alpha'|, \quad (10.12)$$

where $|N, \alpha\rangle$ are the eigenstates of H_s with energies $E_{N\alpha} = \hbar\omega_{N\alpha}$, N being the number of electron-hole pairs and α a collective quantum number denoting the specific electronic level. The state $|N = 0\rangle$ is the electronic ground state, the $N = 1$ subspace is the exciton subspace with the additional collective quantum number α denoting the exciton energy level n , the in-plane wave vector $\mathbf{k}$ and the spin index σ . When needed we will adopt the following notation: $\alpha \equiv (n, k)$ with $k \equiv (\mathbf{k}, \sigma)$. The electronic semiconductor Hamiltonian can be rewritten as

$$\hat{H}_s = \hat{H}_0 + \hat{V}_{\text{Coul}} = \sum_{N\alpha} E_{N\alpha} |N\alpha\rangle \langle N\alpha|. \quad (10.13)$$

In QWs, light and heavy holes in valence band are split off in energy. Assuming that this splitting is much larger than the kinetic energies of all the involved particles and, as well, much larger than the interaction between them, we shall consider only heavy hole states as occupied. As opposed to the bulk case, in a QW single particle states experience confinement along the growth direction and subbands appear, anyway in the other two orthogonal directions translational invariance is preserved and the in-plane exciton wave vector remains a good quantum number. Typically, the energy difference between the lowest QW subband level and the first excited one is larger than the Coulomb interaction between particles, and we will consider excitonic states arising from electrons and heavy holes in the lowest subbands.

The eigenstates of the Hamiltonian $\hat{H}_c$ of the cavity modes can be written as $|n, \lambda\rangle$ where n stands for the total number of photons in the state and $\lambda = (\mathbf{k}_1, \sigma_1; \dots; \mathbf{k}_n, \sigma_n)$ specifies wave vector and polarization σ of each photon. Here

we shall neglect the longitudinal- transverse splitting of polaritons [15] originating mainly from the corresponding splitting of cavity modes. It is more relevant at quite high in-plane wave vectors and often it results to be smaller than the polariton linewidths. The present description can be easily extended to include it. We shall treat the cavity field in the quasi-mode approximation, that is to say we shall quantize the field as the mirrors were perfect and subsequently we shall couple the cavity with a statistical reservoir of a continuum of external modes. This coupling is able to provide the cavity losses as well as the feeding of the coherent external impinging pump beam. The cavity mode Hamiltonian, thus, reads

$$\hat{H}_c = \sum_k \hbar \omega_k^c \hat{a}_k^\dagger \hat{a}_k, \quad (10.14)$$

with the operator $\hat{a}_k^\dagger$ which creates a photon state with energy $\hbar \omega_k^c = \hbar(\omega_{\text{exc}}^2 + v^2|\mathbf{k}|^2)^{1/2}$, v being the velocity of light inside the cavity and $k = (\sigma, \mathbf{k})$. The coupling between the electron system and the cavity modes is given in the usual rotating wave approximation [16, 17]

$$\hat{H}_I = - \sum_{nk} V_{nk}^* \hat{a}_k^\dagger \hat{B}_{nk} + H.c., \quad (10.15)$$

$V_{n,k}$ is the photon-exciton coupling coefficient enhanced by the presence of the cavity [18] set as $V_{n,k} = \tilde{V}_\sigma \sqrt{A} \phi_{n,\sigma}^*(\mathbf{x} = 0)$, the latter being the real-space exciton envelope function calculated in the origin, whereas A is the in-plane quantization surface, $\tilde{V}_\sigma$ is proportional to the interband dipole matrix element. Modelling the loss through the cavity mirrors within the quasi-mode picture means we are dealing with an ensemble of external modes, generally without a particular phase relation among themselves. An input light beam impinging on one of the two cavity mirrors is an external field as well and it must belong to the family of modes of the corresponding side (i.e. left or right). Being coherent, it will be the non zero expectation value of the ensemble. It can be shown [16, 19] that for a coherent input beam, the driving of the cavity modes may be described by the model Hamiltonian [16, 19]

$$\hat{H}_p = it_c \sum_{\mathbf{k}} (E_{\mathbf{k}} \hat{a}_{\mathbf{k}}^\dagger - E_{\mathbf{k}}^* \hat{a}_{\mathbf{k}}), \quad (10.16)$$

where t_c determines the fraction of the field amplitude passing through the cavity mirror, $E_{\mathbf{k}}$ ($E_{\mathbf{k}}^*$) is a $\mathbb{C}$ -number describing the positive (negative) frequency part of the coherent input light field amplitude.

10.3 Linear and nonlinear dynamics

In any theory aiming at describing the dynamics of semiconductor electrons interacting with a light field an infinite hierarchy of dynamical variables is encountered, requiring consideration of an appropriate truncation procedure.

The idea is not to use a density matrix approach, but to derive directly expectation values of all the quantities at play. The dynamics is described by "transition" operators (known as generalized Hubbard operators):

$$\begin{aligned}\hat{X}_{N,\alpha;M,\beta} &= |N, \alpha\rangle \langle M, \beta| \\ \hat{Y}_{n,\lambda;m,\mu} &= |n, \lambda\rangle \langle m, \mu|.\end{aligned}\tag{10.17}$$

The fundamental point in the whole analysis is that, due to the form of the interaction Hamiltonian $\hat{H}_I$ and owing to the quasiparticle conservation property of the free Hamiltonians, we can use the so-called dynamics controlled truncation scheme (DCTS), stating that we are facing a rather special model where the correlation have their origin only in the action of the electromagnetic field and thus the general theorem due to Axt and Stahl [20], and its generalisation to the full quantum case [16], hold. In the cases of coherent optical phenomena the DCTS scheme represents a classification of higher-order density matrices (or dynamical variables) according to their leading order scaling with the applied laser field. It is a rigorous theorem inspired by a classification of nonlinear optical processes [21]. Apart from semiconductors, other systems such as Frenkel exciton systems (e.g. molecular aggregates, molecular crystals or biological antenna systems) are also commonly described in terms of models sharing this property. Indeed, DCTS type approaches have been formulated for these systems too [10]. DCTS can be regarded as a systematic way of selecting a set of relevant dynamical variables, once a given order is set, the truncation is made by neglecting those terms that affect the optical response in orders higher than the given cut-off order. The result is a closed set of coupled nonlinear (with respect to the perturbative parameter) equations that, when solved numerically, give rise to contributions of arbitrarily high orders in the laser field. The main difference from strict perturbation theory is that DCTS expands the equations of motion whereas in perturbation theory the solution is expanded. As a consequence one can say that the range of validity of DCTS includes that of strict perturbation theory, but it is not limited to the latter and resummations up to infinite orders in the fields are implicit in the DCTS equations. Originally the DCTS scheme truncates only the electronic branch of the hierarchy, but very soon it has been clear it has far reaching implementations when combined with other techniques, e.g. projection techniques as those in [11, 16, 17]. Moreover, it can be combined with every strategy to deal with the phonon-assisted branch of the hierarchy [22, 23], or others environment interactions. These relations can be derived by working out the consequences of charge conservation for a system where the carriers are generated only in pairs. The most widely used level of the DCTS theory is obtained by taking the cut for the truncation at the third order and using all available identities for reducing the number of dynamical variables. The DCTS version we will need was conceived in order to include the quantization of the electromagnetic field [16] and allowed the first pioneering works and proposals on genuine quantum effects in optically excited semiconductors [24], experimentally realized only lately [12]. Expressed in our notation it reads [16]:

$$\begin{aligned} \langle \hat{X}_{N,\alpha;M,\beta} \hat{Y}_{n,\lambda;m,\mu} \rangle &= \sum_{i=0}^{i_0} \langle \hat{X}_{N,\alpha;M,\beta} \hat{Y}_{n,\lambda;m,\mu} \rangle^{(N+M+n+m+2i)} \\ &+ \mathcal{O}(E^{(N+M+n+m+2i_0+2)}) \end{aligned} \quad (10.18)$$

the expectation value of a **zero to N-pair** transition is at least of order N in the external field. There are only even powers because of the spatial inversion symmetry which is present.

The exciton and photon operators can be expressed as

$$\begin{aligned} \hat{a}_k &= \hat{Y}_{0;1k} + \sum_{n \geq 1} \sqrt{n_k + 1} \hat{Y}_{n_k; (n_k+1)k} \\ \hat{B}_{nk} &= \hat{X}_{0;1nk} + \sum_{N \geq 1, \alpha\beta} \langle N\alpha | \hat{B}_{nk} | (N+1)\beta \rangle \hat{X}_{N\alpha; (N+1)\beta}, \end{aligned} \quad (10.19)$$

where in writing the photon expansion we omitted all the states not belonging to the k -th mode which add up giving the identity in every Fock sector [25]. In the Heisenberg picture we start considering the equation of motion for the photon and exciton operators, once taken the expectation values we exploit theorem (10.18) retaining only the linear terms. With the help of the transition operators all this procedure may be done by inspection. The linear dynamics for $\langle \hat{a}_k \rangle^{(1)} = \langle \hat{Y}_{0;1nk} \rangle^{(1)}$ and $\langle \hat{B}_{nk} \rangle^{(1)} = \langle \hat{X}_{0;1nk} \rangle^{(1)}$ reads:

$$\frac{d}{dt} \langle \hat{a}_k \rangle^{(1)} = -i\bar{\omega}_k^c \langle \hat{a}_k \rangle^{(1)} + i \sum_n \frac{V_{nk}^*}{\hbar} \langle \hat{B}_{nk} \rangle^{(1)} + t_c \frac{E_k}{\hbar} \quad (10.20)$$

$$\frac{d}{dt} \langle \hat{B}_{nk} \rangle^{(1)} = -i\bar{\omega}_{1nk} \langle \hat{B}_{nk} \rangle^{(1)} + i \frac{V_{nk}}{\hbar} \langle \hat{a}_k \rangle^{(1)}. \quad (10.21)$$

In these equations $\bar{\omega}_k^c = \omega_k^c - i\gamma_k$, where γ_k is the cavity damping, analogously $\bar{\omega}_{1nk} = \omega_{1nk} - i\Gamma_x$ and $\bar{\omega}_{2\beta} = \omega_{2\beta} - i\Gamma_{xx}$, with Γ_x and Γ_{xx} exciton and biexciton damping respectively. The dynamics up to the third order is a little bit more complex (in the following the suffix $^{+(n)}$ stands for "up to" n -th order terms in the external electromagnetic exciting field). After a bit of algebra we obtain [26]

$$\frac{d}{dt} \langle \hat{a}_k \rangle^{+(3)} = -i\bar{\omega}_k^c \langle \hat{a}_k \rangle^{+(3)} + i \sum_n \frac{V_{nk}^*}{\hbar} \langle \hat{B}_{nk} \rangle^{+(3)} + t_c \frac{E_k}{\hbar}, \quad (10.22)$$

$$\begin{aligned} \frac{d}{dt} \langle \hat{B}_{nk} \rangle^{+(3)} &= -i\bar{\omega}_{1nk} \langle \hat{B}_{nk} \rangle^{+(3)} + i \frac{V_{nk}}{\hbar} \langle \hat{a}_k \rangle^{+(3)} \\ &+ \sum_{\tilde{n}\tilde{k}} \left[\frac{i}{\hbar} \sum_{n'k', \alpha} V_{n'k'} \langle 1\tilde{n}\tilde{k} | [\hat{B}_{nk}, \hat{B}_{n'k'}^\dagger] - \delta_{(n'k'); (nk)} | 1\alpha \rangle \langle \hat{X}_{1\tilde{n}\tilde{k}; 1\alpha} \hat{Y}_{0;1k'} \rangle^{(3)} \right. \\ &\quad \left. - i \sum_{\beta} (\omega_{2\beta} - \omega_{1\tilde{n}\tilde{k}} - \omega_{1nk}) \langle 1\tilde{n}\tilde{k} | \hat{B}_{nk} | 2\beta \rangle \langle \hat{X}_{1\tilde{n}\tilde{k}; 2\beta} \hat{Y}_{0;0} \rangle^{(3)} \right], \end{aligned} \quad (10.23)$$

in analogy with Ref. [16] (see also Ref. [11]). The resulting equation of motion for the lowest order biexciton amplitude is

$$\begin{aligned} \frac{d}{dt} \langle \hat{X}_{0,2\beta} \rangle^{(2)} = & -i\bar{\omega}_{2\beta} \langle \hat{X}_{0,2\beta} \rangle^{(2)} \\ & + \frac{i}{\hbar} \sum_{n'k'; n''k''} V_{n'k'; n''k''} \langle 2\beta | \hat{B}_{n'k'}^\dagger | 1n''k'' \rangle \langle \hat{X}_{0,1n''k''} \hat{Y}_{0,1k'} \rangle^{(2)}. \end{aligned} \quad (10.24)$$

Equation 10.23 can be cast in a more useful fashion, by separating the Coulomb contribution into a mean-field interaction between excitons and a genuine four-particle correlation term. This procedure has been developed by Östereich *et al.* [11] in 1995 within a semiclassical approximation. The variable describing two-pair transitions can be eliminated by formally inverting the corresponding equations of motion. This results in a memory kernel representation of the two-pair transitions that is nonlocal in time and enters the equation of motion of single pair transition densities. Although the first theory describing quantum optical effect in exciton systems based on DCTS was developed in 1996 [16], only recently [26] it has been shown that under certain reasonable approximation the equations of motion for the exciton and photon operators can be cast in a form closely resembling those obtained within the semiclassical framework. In this way a unified picture of semiclassical and nonclassical nonlinear optical effects can be achieved. We are addressing a coherent optical response, thus we may consider that a coherent pumping mainly generates *coherent* nonlinear processes, as a consequence the dominant contribution of the biexciton sector on the third-order nonlinear response can be calculated considering the nonlinear term as originating mainly from coherent contributions. In the following we will replace quantum operators at $\mathbf{k} = \mathbf{k}_p$, the pump wave vectors, with classical amplitudes (C-numbers). From the point of view of quantum effects, this approximation implies that nonclassical correlations are taken into account up to the lowest order, namely the standard linearisation procedure of quantum correlations adopted for large systems [27].

The most common set-up for parametric emission is the one where a single coherent pump feed resonantly excites the structure at a given energy and wave vector, $\mathbf{k}_p$. The generalization to multi-pump set-up is straightforward. In order to be more *specific* we shall derive explicitly the case of input light beams activating only the 1S exciton sector with all the same circularly (e.g. σ^+) polarization, thus excluding the coherent excitation of bound two-pair coherences (biexciton) mainly responsible for polarization-mixing [11]. This situation can be realized, for instance, as soon as the biexciton resonance has been carefully tailored off-resonance with respect to the characteristic energies of the states involved in the parametric scattering [28]. We shall show that under this approximation, we end up with a set of coupled equations analogous to those obtained in the semiclassical framework of coherent $\chi^{(3)}$ response (quantized electron system, classical light field) in a QW [11], the main difference being that here the (intracavity) light field is regarded not as a driving external source but as a dynamical field [29]. Nevertheless completely different results

can be obtained for exciton or photon number expectation values or for the higher order correlation functions [24, 17]. This close correspondence for the dynamics of expectation values of the exciton operators, is a consequence of the linearisation of quantum fluctuations. However the present approach includes the light field quantization and can thus be applied to the description of quantum optical phenomena. In addition, having a precise set-up chosen, we will be able to specialize our equations and give an explicit account of the parametric contributions as well as the shifts provided by the lowest order nonlinear dynamics. We start from the Heisenberg equations for the exciton and photon operators with terms providing only lowest order nonlinear response (in the input light field), Eqs. (10.23) and (10.23), we shall retain only the dominant contributions, namely those containing the semiclassical pump amplitude at k_p twice, thus focusing on the "direct" pump-induced nonlinear parametric scattering processes. Following Ref. [26], the lowest order ($\chi^{(3)}$) nonlinear optical response in SMCs is given by the following set of coupled equations:

$$\begin{aligned}\frac{d}{dt}\hat{a}_{\pm\mathbf{k}} &= -i\omega_{\mathbf{k}}^c\hat{a}_{\pm\mathbf{k}} + i\frac{V}{\hbar}\hat{B}_{\pm\mathbf{k}} + t_c\frac{E_{\pm\mathbf{k}}}{\hbar} \\ \frac{d}{dt}\hat{B}_{\pm\mathbf{k}} &= -i\omega_{\mathbf{k}}\hat{B}_{\pm\mathbf{k}} + \hat{s}_{\pm\mathbf{k}} + i\frac{V}{\hbar}\hat{a}_{\pm\mathbf{k}} - \frac{i}{\hbar}R_{\pm\mathbf{k}}^{NL},\end{aligned}\quad (10.25)$$

where $R_{\pm\mathbf{k}}^{NL} = (R_{\pm\mathbf{k}}^{sat} + R_{\pm\mathbf{k}}^{xx})$

$$\begin{aligned}R_{\pm\mathbf{k}}^{sat} &= \frac{V}{n_{\text{sat}}}B_{\pm\mathbf{k}_p}a_{\pm\mathbf{k}_p}\hat{B}_{\pm\mathbf{k}_i}^\dagger \\ R_{\pm\mathbf{k}}^{xx} &= \hat{B}_{\pm\mathbf{k}_i}^\dagger(t)\left(V_{xx}B_{\pm\mathbf{k}_p}(t)B_{\pm\mathbf{k}_p}(t) \right. \\ &\quad \left. - i\int_{-\infty}^t dt' F^{\pm\pm}(t-t')B_{\pm\mathbf{k}_p}(t')B_{\pm\mathbf{k}_p}(t')\right).\end{aligned}\quad (10.26)$$

The pump induced renormalization of the exciton dispersion gives a frequency shift

$$\begin{aligned}\hat{s}_{\pm\mathbf{k}} &= -i\left(\frac{V}{\hbar n_{\text{sat}}}(B_{\pm\mathbf{k}_p}^*a_{\pm\mathbf{k}_p}\hat{B}_{\pm\mathbf{k}} + B_{\pm\mathbf{k}_p}^*B_{\pm\mathbf{k}_p}\hat{a}_{\pm\mathbf{k}}) \right. \\ &\quad \left. + 2\frac{V_{xx}}{\hbar}B_{\pm\mathbf{k}_p}^*B_{\pm\mathbf{k}_p}\hat{B}_{\pm\mathbf{k}} \right. \\ &\quad \left. - 2\frac{i}{\hbar}B_{\pm\mathbf{k}_p}^*(t)\int_{-\infty}^t dt' F^{\pm\pm}(t-t')\hat{B}_{\pm\mathbf{k}}(t')B_{\pm\mathbf{k}_p}(t')\right),\end{aligned}\quad (10.27)$$

where

$$n_{\text{sat}} = \left(\frac{2}{\sqrt{A}\phi^*(0)}O^{\text{PSF}}\right)^{-1} = \frac{A}{\pi a_x^2} \frac{7}{16}, \quad (10.29)$$

A being the quantization surface, $\Phi_{\mathbf{q}} = \frac{1}{\sqrt{A}} \frac{\sqrt{2\pi}a_x}{(1+(a_x|\mathbf{q}|)^2)^{3/2}}$, where a_x is the exciton Bohr radius (notice that $\sum_{\mathbf{q}=-\infty}^{\infty} |\Phi_{\mathbf{q}}|^2 = 1$, and $\phi_{n,\sigma}^*(\mathbf{x}=0)$ the real-space exciton envelope function calculated in the origin).

$$\begin{aligned} V_{xx} &= \frac{\hbar}{2} \langle 0 | \hat{D}_{\bar{k},k} \hat{B}_{k'}^\dagger \hat{B}_{k''}^\dagger | 0 \rangle \\ F_{\bar{k},k}^{k'',k'}(t-t') &= \frac{\hbar}{2} \langle 0 | \hat{D}_{\bar{k},k}(t-t') \hat{D}_{k'',k'}^\dagger | 0 \rangle \\ \hat{D}_{\bar{k},k} &\doteq [\hat{B}_{\bar{k}}, [\hat{B}_k, \hat{H}_c]], \end{aligned} \quad (10.30)$$

where a *force* operator $\hat{D}$ is defined [11]. In order to lighten the notation, we dropped the two spin indexes σ and $\tilde{\sigma}$ in the four-particle kernel function F defined in Eq. (10.30) for they are already univocally determined once chosen the others (i.e. σ' and σ'') as soon as their selection rule ($\delta_{\sigma+\tilde{\sigma};\sigma'+\sigma''}$) is applied. In the specific case under analysis we are considering co-circularly polarized waves and more explicit calculations of the nonlinear Coulomb term in Eq. (10.30) can be found in Refs. [30, 31]. The generalization to arbitrary polarization can be derived translating the semiclassical derivation of Ref. [32] within this full quantum framework

For the range of $\mathbf{k}$ -space of interest, i.e. $|\mathbf{k}| \ll \frac{\pi}{a_x}$ (much lower than the inverse of the exciton Bohr radius) they are largely independent on the center of mass wave vectors. While V_{xx} and $F^{\pm\pm}(t-t')$ (i.e. co-circularly polarized waves) conserve the polarizations, $F^{\pm\mp}(t-t')$ and $F^{\mp\pm}(t-t')$ (counter-circular polarization) give rise to a mixing between the two circular polarizations. The physical origin of the terms in Eq. (10.30) can be easily understood: the first is the Hartee-Fock or mean-field term representing the first order treatment in the Coulomb interaction between excitons, the second term is a pure biexciton (four-particle correlations) contribution. This coherent memory may be thought as a non-Markovian process involving the two-particle (excitons) states interacting with a bath of four-particle correlations [11].

The strong exciton-photon coupling does not modify the memory kernel because four-particle correlations do not couple directly to cavity photons. As pointed out clearly in Ref. [29], cavity effects alter the phase dynamics of two-particle states during collisions, indeed, the phase of two-particle states in SMCs oscillates with a frequency which is modified with respect to that of excitons in bare QWs, thus producing a modification of the integral in Eq. (10.26). This way the exciton-photon coupling V_{nk} affects the exciton-exciton collisions that govern the polariton amplification process. Ref. [29] considers the first (mean-field) and the second (four-particle correlation) terms in the particular case of co-circularly polarized waves, calling them without indexes as V_{xx} and $F(t)$ respectively. Fig. 10.1 shows $\mathcal{F}(\omega)$, the Fourier transform of $F(t)$ plus the mean-field term V_{xx} ,

$$\mathcal{F}(\omega) = V_{xx} - i \int_{-\infty}^{\infty} dt F(t) e^{i\omega t}. \quad (10.31)$$

Its imaginary part is responsible for the frequency dependent excitation induced dephasing, it reflects the density of states of two-exciton pair coherences. Towards the

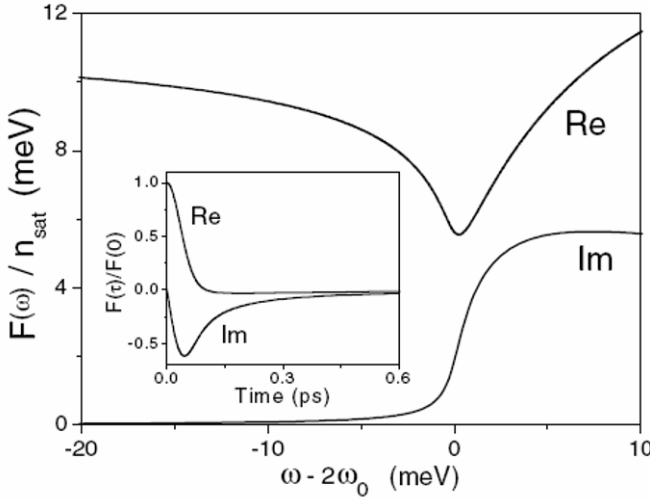


Fig. 10.1 From figure 1 of Ref. [29]. Energy dependence of the effective exciton-exciton scattering potential $\mathcal{F}(\omega)$, calculated for a 7 nm wide GaAs QW with an exciton binding energy of 13.5 meV using a T-matrix approach.

negative detuning region the dispersive part $\text{Re}(\mathcal{F})$ increases whereas the absorptive part $\text{Im}(\mathcal{F})$ goes to zero. The former comprises the mean-field contribution effectively reduced by the four-particle contribution. Indeed, the figure shows the case with a binding energy of 13.5 meV, it gives $V_{xx}n_{\text{sat}} \simeq 11.39$ meV which clearly is an upper bound for $\text{Re}(\mathcal{F})$ for negative detuning. The contribution carried by $F(t)$ determines an effective reduction of the mean-field interaction (through its imaginary part which adds up to V_{xx}) and an excitation induced dephasing. It has been shown [29] that both effects depends on the sum of the energies of the scattered polariton pairs. Equations (10.25) are the main result of the present section. They can be considered the starting point for the microscopic description of quantum optical effects in SMCs. These equations extend the usual semiclassical description of Coulomb interaction effects, in terms of a mean-field term plus a genuine non-instantaneous four-particle correlation, to quantum optical effects. Starting from here, in the strong coupling case, it might be useful to transform the description into a polariton basis. As a consistency check, as soon as we take the expectation values of eqs. (10.25) we obtain a result analogous to the corresponding equations describing the semiclassical (quantized electron system, classical light field) coherent $\chi^{(3)}$ response in a QW [11], the main difference being that here the (intracavity) light field is regarded not as a driving external source but as a dynamical field [29]. This close correspondence for the dynamics of expectation values of the exciton operators, is a consequence of

the linearization of quantum fluctuations. Nevertheless completely different results can be obtained for exciton or photon number expectation values or for higher order correlation function [24, 17]. Moreover, the present approach includes the light field quantization and can thus be applied to the description of quantum optical phenomena.

10.4 Entangled photon pairs from the optical decay of biexcitons

Transient or frequency-resolved four-wave mixing (FWM) are among the most widely used techniques for probing the optical properties of electronic excitations in semiconductors. The FWM process can be schematically described as follows: two incident pump photons with a given wave vector $\mathbf{k}_p$ propagating inside the crystal slab (or the tailored semiconductor structure) as excitonic polaritons, excite a virtual state with two electron-hole pairs with total wave vector $2\mathbf{k}_p$, while a probe beam with wave vector $\mathbf{k}_1$ stimulates the optical decay of the state with two e-h pairs into a final polariton pair at $\mathbf{k}_1$ and $\mathbf{k}_2 = 2\mathbf{k}_p - \mathbf{k}_1$. The states with two e-h pairs can be bound biexcitons or two exciton scattering states. This stimulated process thus determines the generation of a new beam at $\mathbf{k}_2$ as well as the amplification of the probe beam at $\mathbf{k}_1$ (parametric gain) as observed in SMCs. If the pump beam is maintained while eliminating the probe beam, there is no coherent emission at $\mathbf{k}_2$ according to the semiclassical theory. Actually the quantum fluctuations of the light field can play the role of the probe beam *stimulating* the optical decay of states with two e-h pairs into a pair of final polariton states. In particular quantum fluctuations of modes at a generic wave vector $\mathbf{k}_1$ can determine light emission in the direction $\mathbf{k}_2 = 2\mathbf{k}_p - \mathbf{k}_1$ and vice-versa.

The process that we are going to describe is known as hyper-Raman scattering (HRS) or two-photon Raman scattering. HRS can be schematically described as follows (see fig. 10.2): A first photon induces a transition from the ground state to the excitonic subspace, the second photon induces a further transition from the exciton to the biexciton subspace, then the excited biexciton may decay spontaneously into an exciton and a photon such that total energy and wave vectors are preserved. However, we have just mentioned that the allowed propagating modes inside the crystal slab are polaritons. The above picture should be modified accordingly: a pair of incident photons of given wavevector and energy propagate inside the crystal as polaritons of given energy $\hbar\omega(\mathbf{k}_i)$ and may give rise (according to the hierarchy of dynamic equations) to a biexciton excitation, such excitation can finally decay into a pair of final polaritons. Total energy and momentum are conserved in the whole process :

$$\omega(\mathbf{k}_1) + \omega(\mathbf{k}_2) = 2\omega(\mathbf{k}_i),$$

$$\mathbf{k}_1 + \mathbf{k}_2 = 2\mathbf{k}_i,$$

where final polariton states have been labelled with 1 and 2. The determination of the polariton dispersion has successfully been accomplished by HRS in several

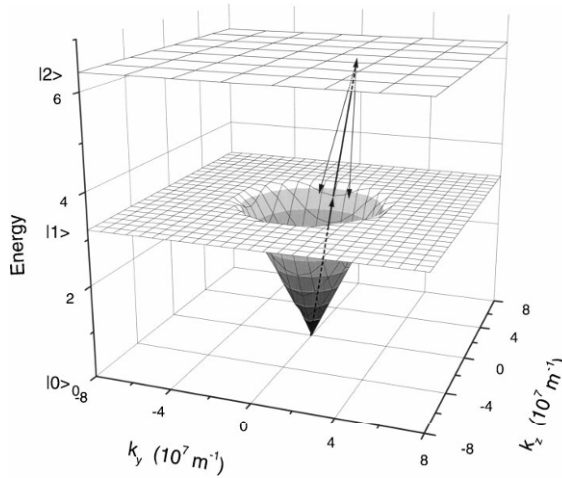


Fig. 10.2 Resonant two-photon Raman scattering process in CuCl involving the lower polariton branch, $|0\rangle, |1\rangle, |2\rangle$ are, respectively, the ground state, the 1s exciton stat and the bound biexciton state.

large-gap bulk semiconductors. HRS is a process related to the third-order nonlinear susceptibility and can be considered as a spontaneous resonant non-degenerate four-wave mixing (FWM). In FWM the optical decay of the coherently excited biexcitons is stimulated by sending an additional light beam, while in HRS the decay is determined by intrinsic quantum fluctuations. This subtle difference imply that HRS is a quantum optical process that cannot be described properly without including the quantization of the light field [33]. The scattering process rate largely increases when a bound biexciton state is resonantly excited by the two polariton process. The final result of the scattering process is the generation of polaritons with slightly different wavevectors and energy displaying pair quantum correlations. If the two final quanta are polaritons with a significant photon component, at the end of the slab they can escape the semiconductor as photons and can be both detected. Since the bound biexciton is formed by carriers with oppisite spin, the resulting polariton pairs will be composed by polaritons of opposite spin, whic will give rise to photons with opposite circular polarization, corresponding to the following polarization entangled quantum state:

$$|\Psi\rangle = \frac{1}{\sqrt{2}} (|+\rangle_1 |-\rangle_2 + |-\rangle_1 |+\rangle_2) . \quad (10.32)$$

In 2004, experimental evidence for the generation of ultraviolet polarization-entangled photon pairs by means of biexciton resonant parametric emisison in a single crystal of semiconductor CuCl has been reported [12].

10.5 The picture of interacting polaritons

When the coupling rate V exceeds the decay rate of the exciton coherence and of the cavity field, the system enters the strong coupling regime. In this regime, the continuous exchange of energy before decay significantly alters the dynamics and hence the resulting resonances of the coupled system with respect to those of bare excitons and cavity photons. As a consequence, cavity-polaritons arise as the two-dimensional eigenstates of the coupled system. The coupling rate V determines the splitting ($\simeq 2V$) between the two polariton energy bands. This nonperturbative dynamics including the interactions (induced by $\hat{R}_k^{NL}$) between different polariton modes can be accurately described by Eq. (10.25). Nevertheless there can be reasons to prefer a change of basis from excitons and photons to the eigenstates of the coupled system, namely polaritons. An interesting one is that the resulting equations may provide a more intuitive description of nonlinear optical processes in terms of interacting polaritons. Moreover equations describing the nonlinear interactions between polaritons become more similar to those describing parametric interactions between photons widely adopted in quantum optics. Another, more fundamental reason, is that the standard second-order Born-Markov approximation scheme, usually adopted to describe the interaction with environment, is strongly bases-dependent, and using the eigenstates of the closed system provides more accurate results. Equation (10.25) can be written in compact form as

$$\dot{\mathcal{B}}_k = -i\Omega_k^{xc} \mathcal{B}_k + \mathcal{E}_k^{in} - i\mathcal{R}_k^{NL}; \quad (10.33)$$

where $\mathcal{B}_k \equiv \begin{pmatrix} \hat{B}_k \\ \hat{a}_k \end{pmatrix}$, $\Omega_k^{xc} \equiv \begin{pmatrix} \bar{\omega}_k^x & -V \\ -V & \bar{\omega}_k^c \end{pmatrix}$, $\mathcal{E}_k^{in} \equiv \begin{pmatrix} 0 \\ t_c E_k^{in} \end{pmatrix}$, and $\mathcal{R}_k^{NL} \equiv \begin{pmatrix} \hat{R}_k^{NL} \\ 0 \end{pmatrix}$.

In order to obtain the dynamics for the polariton system we perform unitary basis transformation on the exciton and photon operators

$$\mathcal{P}_k = U_k \mathcal{B}_k; \quad (10.34)$$

being $\mathcal{P}_k = \begin{pmatrix} \hat{P}_{1k} \\ \hat{P}_{2k} \end{pmatrix}$ and

$$U_k = \begin{pmatrix} X_{1k} & C_{1k} \\ X_{2k} & C_{2k} \end{pmatrix}, \quad (10.35)$$

In general photon operators obey Bose statistics, on the contrary the excitons do not possess a definite statistics (i.e. either bosonic or fermionic), but their behaviour may be well approximated by a bosonic-like statistics in the limit of low excitation densities. Indeed

$$[\hat{B}_n, \hat{B}_{n'}^\dagger] = \delta_{n',n} - \sum_{\mathbf{q}} \Phi_{n\mathbf{q}}^* \Phi_{n'\mathbf{q}} \sum_{N,\alpha,\beta} \left(\langle N\alpha | \hat{c}_{\mathbf{q}}^\dagger c_{\mathbf{q}} | N\beta \rangle \right. \\ \left. + \langle N\alpha | \hat{d}_{-\mathbf{q}}^\dagger d_{-\mathbf{q}} | N\beta \rangle \right) | N\alpha \rangle \langle N\beta |, \quad (10.36)$$

Thus, within a DCTS line of reasoning [11], the expectation values of these transition operators (i.e. $|N\alpha\rangle\langle N\beta|$) are at least of the second order in the incident light field simply because they are density-dependent contributions. Evidently all these considerations affect polariton statistics as well, polariton being a linear combination of intracavity photons and excitons. As a consequence, even if polariton operators have no definite statistics, in the limit of low excitation intensities, at first order in the incident field, they obey approximately bosonic-like commutation rules. Diagonalizing $\Omega_{\mathbf{k}}^{\text{xc}}$:

$$U_{\mathbf{k}} \Omega_{\mathbf{k}}^{\text{xc}} = \tilde{\Omega}_{\mathbf{k}} U_{\mathbf{k}}, \quad (10.37)$$

where

$$\tilde{\Omega}_{\mathbf{k}} = \begin{pmatrix} \omega_{1\mathbf{k}} & 0 \\ 0 & \omega_{2\mathbf{k}} \end{pmatrix}.$$

$\omega_{1,2}$ are the eigenenergy (as a function of $\mathbf{k}$) of the lower (1) and upper (2) polariton states. After simple algebra it is possible to obtain this relation for the Hopfield coefficients [34]:

$$X_{1\mathbf{k}} = -C_{2\mathbf{k}}^*; \quad C_{1\mathbf{k}} = X_{2\mathbf{k}}^*. \quad (10.38)$$

where

$$X_{1\mathbf{k}} = \frac{1}{\sqrt{1 + \left(\frac{V}{\omega_{1\mathbf{k}} - \omega_k^c} \right)^2}} \quad C_{1\mathbf{k}} = \frac{1}{\sqrt{1 + \left(\frac{\omega_{1\mathbf{k}} - \omega_k^c}{V} \right)^2}}. \quad (10.39)$$

Introducing this transformation into Eq. (10.33), one obtains

$$\dot{\mathcal{P}}_{\mathbf{k}} = -i\tilde{\Omega}_{\mathbf{k}} \mathcal{P}_{\mathbf{k}} + \tilde{\mathcal{E}}_{\mathbf{k}}^{\text{in}} - i\tilde{\mathcal{R}}_{\mathbf{k}}^{\text{NL}}; \quad (10.40)$$

where $\tilde{\mathcal{R}}^{\text{NL}} = U \mathcal{R}^{\text{NL}}$, which in explicit form reads

$$\dot{\hat{P}}_{1\mathbf{k}} = -i\omega_{1\mathbf{k}} \hat{P}_{1\mathbf{k}} - i\tilde{s}_{1\mathbf{k}} + \tilde{E}_{1,\mathbf{k}}^{\text{in}} - i\tilde{R}_{1\mathbf{k}}^{\text{NL}}, \quad (10.41a)$$

$$\dot{\hat{P}}_{2\mathbf{k}} = -i\omega_{2\mathbf{k}} \hat{P}_{2\mathbf{k}} - i\tilde{s}_{2\mathbf{k}} + \tilde{E}_{2,\mathbf{k}}^{\text{in}} - i\tilde{R}_{2\mathbf{k}}^{\text{NL}}; \quad (10.41b)$$

where $\tilde{E}_{m\mathbf{k}}^{\text{in}} = t_c C_{m\mathbf{k}} E_{\mathbf{k}}^{\text{in}}$, and $\tilde{R}_{m\mathbf{k}}^{\text{NL}} = X_{i\mathbf{k}} \hat{R}_{\mathbf{k}}^{\text{NL}}$, ($m = 1, 2$). Such a diagonalization is the necessary step when the eigenstates of the polariton system are to be used used as starting states perturbed by the interaction with the environment degrees of freedom [35]. The nonlinear interaction written in terms of polariton operators reads

$$\hat{R}_{\mathbf{k}}^{NL} = \sum_{i,j,l} \hat{P}_{i\mathbf{k}_i}^\dagger(t) \int_{-\infty}^t g_{m\mathbf{k}}^{ijl}(t,t') P_{j\mathbf{k}_p}(t') P_{l\mathbf{k}_p}(t') dt', \quad (10.42)$$

where

$$g_{m\mathbf{k}}^{ijl}(t,t') = \frac{1}{N_{eff}} \left[\frac{V}{n_{sat}} C_{j,\mathbf{k}_p}^* \delta(t-t') \right. \\ \left. + \left(V_{xx} \delta(t-t') - iF(t-t') \right) X_{j,\mathbf{k}_p}^* \right] X_{l,\mathbf{k}_p}^* X_{i,\mathbf{k}_i}. \quad (10.43)$$

N_{eff} depends on the number of wells inside the cavity and their spatial overlap with the cavity mode. The shift $\hat{s}_{\mathbf{k}}(t)$ is transformed into

$$\tilde{s}_{m\mathbf{k}}(t) = \sum_{ijl} P_{i\mathbf{k}_p}^*(t) \int_{-\infty}^t \left(h_{m\mathbf{k}}^{ijl} \delta(t-t') - 2iF(t-t') \right) P_{j\mathbf{k}_p}(t') \hat{P}_{l\mathbf{k}}(t') dt', \quad (10.44)$$

and

$$h_{m\mathbf{k}}^{ijl} = \frac{1}{N_{eff}} X_{m\mathbf{k}} \left[\frac{V}{n_{sat}} X_{i\mathbf{k}_p} \left(C_{j,\mathbf{k}_p}^* X_{l\mathbf{k}}^* + X_{j\mathbf{k}_p}^* C_{l\mathbf{k}}^* \right) \right. \\ \left. + 2V_{xx} X_{i\mathbf{k}_p} X_{j\mathbf{k}_p}^* X_{l\mathbf{k}}^* \right]. \quad (10.45)$$

Equation (10.41) describes the coherent dynamics of a system of interacting cavity polaritons. The nonlinear term drives the mixing between polariton modes with different in-plane wave vectors and possibly belonging to different branches.

Analogous equations can be obtained starting from an effective Hamiltonian describing excitons as interacting bosons [36]. The resulting equations (usually developed in a polariton basis) do not include correlation effects beyond Hartree-Fock. Moreover the interaction terms due to phase space filling differs from those obtained within the present approach, not based on an effective Hamiltonian. Indeed, eqs. (10.40) have nonlinear terms of the same structure of Ref. [36] (see eqs.(43-44) even if they are already written in the polariton basis), but display two main differences originating from the different starting points. Our equations, obtained from the DCTS, includes the non-instantaneous four-particle correlation determining a correction to the mean-field Coulomb interaction and a frequency-dependent excitation-induced dephasing. Moreover, whereas the mean-field Coulomb interaction coincides in the two approaches, the interaction term originating from phase space filling differs.

In particular (for $N_{eff} = 1$, considering only lower polaritons, and for real Hopfield coefficients) we obtain

$$R_{\mathbf{k}}^{NL}|_{psf} = \frac{V}{n_{sat}} \sum_{\mathbf{k}',\mathbf{q}} C_{\mathbf{k}'-\mathbf{q}} X_{\mathbf{k}+\mathbf{q}} X_{\mathbf{k}'} P_{\mathbf{k}'}^* P_{\mathbf{k}+\mathbf{q}} P_{\mathbf{k}'-\mathbf{q}}. \quad (10.46)$$

the corresponding term in [36] can be written as

$$\begin{aligned}
R_{\mathbf{k}}^{NL}|_{psf} = & 2 \frac{V}{n_{sat}} \sum_{\mathbf{k}', \mathbf{q}} C_{\mathbf{k}'-\mathbf{q}} X_{\mathbf{k}+\mathbf{q}} X_{\mathbf{k}'} P_{\mathbf{k}'}^* P_{\mathbf{k}+\mathbf{q}} P_{\mathbf{k}'-\mathbf{q}} \\
& + \frac{V}{n_{sat}} \sum_{\mathbf{k}', \mathbf{q}} C_{\mathbf{k}'} X_{\mathbf{k}+\mathbf{q}} X_{\mathbf{k}'-\mathbf{q}} P_{\mathbf{k}'}^* P_{\mathbf{k}+\mathbf{q}} P_{\mathbf{k}'-\mathbf{q}}
\end{aligned} \quad (10.47)$$

contains additional terms providing a larger (by a factor 3) interaction strength due to phase space filling and displaying a different k -dependence. We believe that the difference is mainly due to the adopted Bosonization procedure. According to that procedure the exciton operator (determining the resonant polarization) $\hat{B}$ is expanded in terms of Bose operators $\hat{B}_B$ up to the first two terms. Schematically $\hat{B} \rightarrow \hat{B}_B + \hat{B}_B^\dagger \hat{B}_B \hat{B}_B + \dots$. Then the equation of motion for $\langle \hat{B}_B \rangle$ is obtained. The discrepancy may arise from the fact that $\hat{B}_B + \hat{B}_B^\dagger \hat{B}_B \hat{B}_B$ and not $\hat{B}_B$ should be regarded as the proper polarization operator. It is worth noting that more rigorously calculated nonlinear coupling coefficients will describe more accurately parametric dynamics, as evidenced by the good quantitative agreement with experimental data, our numerical results show (see Fig.) [19, 37], where we tested numerically our framework. Only the many-body electronic Hamiltonian, the intracavity-photon Hamiltonian and the Hamiltonian describing their mutual interaction have been taken into account. Losses through mirrors, decoherence and noise due to environment interactions as well as applications of this theoretical framework, in the strong coupling regime, will be addressed in the next sections.

10.6 Noise and environment: Quantum Langevin approach

Polaritons are mixed quasiparticles resulting from the strongly coupled propagation of light and collective electronic excitations (excitons) in semiconductor crystals. Although spontaneous parametric processes involving polaritons in bulk semiconductors have been known for decades[38], the possibility of generating entangled photons by these processes was theoretically pointed out only lately[16]. This result was based on a microscopic quantum theory of the nonlinear optical response of interacting electron systems relying on the DCTS [39] extended to include light quantization[17, 24, 29]. The above theoretical framework was also applied to the analysis of polariton parametric emission in semiconductor microcavities (SMCs)[17, 29].

However they are real electronic excitations propagating in a complex interacting environment. Owing to the relevance of polariton interactions, and also owing to their interest for exploring quantum optical phenomena in such a complex environment, theoretical approaches able to model accurately polariton dynamics including light quantization, losses and environment interactions are highly desired. The analysis of nonclassical correlations in semiconductors constitutes a challenging problem, where the physics of interacting electrons must be added to quantum

optics and should include properly the effects of energy relaxation, dephasing, and noise, induced by electron-phonon interaction [14].

Previous descriptions of polariton parametric processes make deeply use of the picture of polaritons as interacting bosons. These theories have been used to investigate parametric amplifications, parametric luminescence, coherent control, entanglement and parametric scattering in momentum space[36, 40, 41, 42, 43].

It is worth noting that in a realistic environment phase-coherent nonlinear optical processes involving real excitations compete with incoherent scattering as evidenced by experimental results. In experiments dealing with parametric emission, what really dominates emission at low pump intensities is the photoluminescence (PL) due to the incoherent dynamics of the single scattering events driven by the pump itself and the Rayleigh scattering of the pump due to the unavoidable presence of structural disorder. The latter process is elastic and can thus be spectrally filtered in principle, moreover it is confined in k -space to a ring of in-plane wave vectors with almost the same modulus of the pump wave vector. On the contrary PL, being not an elastic process, cannot be easily separated from parametric emission. Only when the pumping becomes sufficient the parametric processes start to reveal themselves and to take over pump-induced PL as well. Indeed, parametric emission and standard pump-induced PL usually coexist as shown by experiments at low and intermediate excitation density[43]. Moreover, in order to address quantum coherence properties and entanglement[12] the preferred experimental situations are those of few-particle regimes, namely coincidence detection in photon counting. In this regime, the presence of incoherent noise due to pump-induced PL tends to spoil the system of its coherence properties lowering the degree of nonclassical correlations. The detrimental influence of incoherent effects on the quantum coherence properties is also well evidenced in the measured time-resolved visibility shown in Ref. [13]. At initial times visibility is suppressed until parametric emission prevails. Thus, a microscopic analysis able to account for parametric emission and pump-induced PL on an equal footing is highly desirable in order to make quantitative comparison with measurements and propose future experiments. Furthermore a quantitative theory would be of paramount importance for a deeper understanding of quantum correlations in such structures aiming at seeking and limiting all unwanted detrimental contributions.

In order to model the quantum dynamics of the polariton system in the presence of losses and decoherence we exploit the microscopic quantum Heisenberg-Langevin approach. We choose it because of its easiness in manipulating operator differential equations, and above all, for its invaluable flexibility and strength in performing even multi-time correlation calculations, that are particularly important when dealing with quantum correlation properties of the emitted light. Moreover, as we shall see in the following, it enables, under certain assumptions, a (computationally advantageous) decoupling of incoherent dynamics from parametric processes.

In the standard well-known theory of quantum Langevin noise treatment [44] greatly exploited in quantum optics, one uses a perturbative description and thanks to a Markov approximation gathers the damping as well as a term including the correlation of the system with the environment. The latter arises from the initial values

of the bath operators, which are assumed to behave as noise sources of stochastic nature. Normally the model considered has the form of harmonic oscillators coupled linearly to a bosonic environment. The standard statistical viewpoint is easily understood: the unknown initial values of the bath operators are considered as responsible for fluctuations, and the most intuitive idea is to assume bosonic commutation relations for the Langevin noise sources because the bath is bosonic too. Most times these commutation relations are introduced phenomenologically with damping terms taken from experiments and/or from previous works. In other contexts a microscopic calculation has been attempted using a quantum operator approach. Besides its valuable results as soon as one tries to set a microscopic calculation for interaction forms different from a 2-body linear coupling [44], e.g. acoustic-phonon interaction, some problems arise and one is forced to consider additional approximations in order to close the equations of motion and obtain damping and fluctuations.

In 1966 Melvin Lax, having in mind the lesson of classical statistical mechanics of Brownian motion, extended the noise-source technique to quantum systems. In general, the model comprises a system of interest coupled to a reservoir (R). Considering a generic global (i.e. system+reservoirs) operator, a first partial trace over the reservoir degrees of freedom results still in a system operator, a subsequent trace over the system degrees of freedom would give an expectation value. In order to be as clear as possible we shall denote the former operation on the environment by single brackets $\langle \rangle_R$, whereas for the combination of the two (partial trace over the reservoir and subsequent partial trace over the system density matrices) the usual brackets $\langle \rangle$ are used. His philosophy was that the reservoir can be completely eliminated provided that frequency shift and dissipation induced by the reservoir interactions are incorporated into the mean equations of motion, and provided that suitable operator noise sources with the correct moments are added. In Ref. [45] he proposed for the first time that as soon as one is left with a closed set of equations of system operators for the mean motion (*mean* with respect to the reservoir) they can be promoted to equations for global bare operators (system+reservoir) provided additive noise sources endowed by the proper statistics due to the system dynamics are considered. He showed that in a Markovian environment these noise source operators must fulfill generalized Einstein equations which might be considered as an alternative form of time-dependent non-equilibrium fluctuation-dissipation theorem. If $\hat{\mathbf{a}} = \{\hat{a}_1, \hat{a}_2, \dots\}$ is a set of system operators, and

$$\frac{d\langle \hat{a}_\mu \rangle_R}{dt} = \langle \hat{A}_\mu(\hat{\mathbf{a}}) \rangle_R \quad (10.48)$$

are the correct equations for the mean, then one can show that the equations

$$\frac{d\hat{a}_\mu}{dt} = \hat{A}_\mu(\hat{\mathbf{a}}) + \hat{F}_\mu(\hat{\mathbf{a}}, t) \quad (10.49)$$

are a valid set of equations of motion for the operators provided the additive noise operators $\hat{F}$'s to be possess the correct statistical properties to be determined for the motion itself.

The Langevin noise source operators are such that their expectation values $\langle \hat{\mathcal{F}}_\mu \rangle_R$ vanish, but their second order moments do not [45]. They are intimately linked up with the global dissipation and in a Markovian environment they take the form:

$$\langle \hat{\mathcal{F}}_\mu(t) \hat{\mathcal{F}}_\nu(u) \rangle_R = 2 \langle \hat{D}_{\mu\nu} \rangle_R \delta(t-u), \quad (10.50)$$

where the diffusion coefficients are

$$2 \langle \hat{D}_{\mu\nu} \rangle_R = \frac{d}{dt} \langle \hat{a}_\mu(t) \hat{a}_\nu(t) \rangle_R - \left\langle \left\{ \frac{d}{dt} \hat{a}_\mu \right\} \hat{a}_\nu \right\rangle_R - \langle \hat{a}_\mu \left\{ \frac{d}{dt} \hat{a}_\nu \right\} \rangle_R, \quad (10.51)$$

$$\left\{ \frac{d}{dt} \hat{a}_\nu \right\} \equiv \frac{d}{dt} \hat{a}_\nu - \hat{\mathcal{F}}_\nu. \quad (10.52)$$

Equation (10.50) is an (exact) time dependent Einstein equation representing a fluctuation-dissipation relation valid for nonequilibrium situations, it reflects the fundamental correspondence between dissipation and noise in an open system. $\langle \hat{D}_{\mu\nu} \rangle_R$ becomes not only time-dependent, it is a system operator and can be seen as the extent to which the usual rules for differentiating a product is violated in a Markovian system. Equation (10.50) and Eq. (10.51) make the resulting "fluctuation-dissipation" relations between $\hat{D}_{\mu\nu}$ and the reservoir contributions to be in precise agreement with those found by direct use of perturbation theory. This method, however, guarantees the commutation rules for the corresponding operators to be necessarily preserved in time. This result is more properly an exact, quantitative theorem which gives relevant insights regarding the intertwined microscopic essence of damping and fluctuations in any open system.

In order to be more *specific*, let us consider a single semiclassical pump feed resonantly exciting the lower polariton branch at a given wave vector $\mathbf{k}_p$. It is worth noting, however, that the generalization to a many-classical-pumps settings is straightforward. The nonlinear term R^{NL} of Eq. (10.42) couples pairs of wave vectors, let's say $\mathbf{k}$, the signal, and $\mathbf{k}_i = 2\mathbf{k}_p - \mathbf{k}$, the idler. The Heisenberg Eqs. (10.41), involving system operators, for the generic couple read

$$\begin{aligned} \frac{d}{dt} \langle \hat{P}_{\mathbf{k}} \rangle_R &= -i\tilde{\omega}_{\mathbf{k}} \langle \hat{P}_{\mathbf{k}} \rangle_R + g_{\mathbf{k}} \langle \hat{P}_{\mathbf{k}_i}^\dagger \rangle_R \mathcal{P}_{\mathbf{k}_p}^2 \\ \frac{d}{dt} \langle \hat{P}_{\mathbf{k}_i}^\dagger \rangle_R &= i\tilde{\omega}_{\mathbf{k}_i} \langle \hat{P}_{\mathbf{k}_i}^\dagger \rangle_R + g_{\mathbf{k}_i}^* \langle \hat{P}_{\mathbf{k}} \rangle_R \mathcal{P}_{\mathbf{k}_p}^2, \end{aligned} \quad (10.53)$$

where we changed slightly the notation to underline that pump polariton amplitudes $\mathcal{P}_{\mathbf{k}_p}$ are regarded as classical variables ($\mathbb{C}$ -numbers), while the generated signal and idler polaritons are regarded as true quantum variables.

The nonlinear interaction terms in Eq. (10.53) reads

$$g_{\mathbf{k}} = \frac{1}{N_{eff}} \left[\frac{V}{n_{sat}} C_{\mathbf{k}_p}^* + V_{xx} X_{\mathbf{k}_p}^* \right] X_{\mathbf{k}_p} X_{\mathbf{k}_i}^* X_{\mathbf{k}_i}. \quad (10.54)$$

It accounts for a pump-induced blue-shift of the polariton resonances and a pump-induced parametric emission. In Eqs. (10.53) only nonlinear terms arising from saturation and from the mean-field Coulomb interaction have been included. Correlation effects beyond mean-field introduce non-instantaneous nonlinear terms. They mainly determine an effective reduction of the mean-field interaction and an excitation induced dephasing. It has been shown [29] that both effects depend on the sum of the energies of the scattered polariton pairs. While the effective reduction can be taken into account simply modifying V_{xx} , the proper inclusion of the excitation induced dephasing requires the explicit inclusion into the dynamics of four-particle states with their phonon-induced scattering and relaxation. In the following we will neglect this effect that is quite low at zero and even less at negative detuning on the lower polariton branch [46]. The renormalized complex polariton dispersion $\tilde{\omega}_{\mathbf{k}}$ includes the effects of relaxation and pump-induced renormalization, $\tilde{\omega}_{\mathbf{k}} = \omega_{\mathbf{k}} - i\Gamma_{\mathbf{k}}/2 + h_{\mathbf{k}} |\mathcal{P}_{\mathbf{k}_p}|^2$, and

$$h_{\mathbf{k}} = \frac{1}{N_{eff}} \left(\frac{V}{n_{sat}} C_{\mathbf{k}_p}^* X_{\mathbf{k}_p} |X_{\mathbf{k}}|^2 + \frac{V}{n_{sat}} C_{\mathbf{k}}^* X_{\mathbf{k}} |X_{\mathbf{k}_p}|^2 + 2V_{xx} |X_{\mathbf{k}_p}|^2 |X_{\mathbf{k}}|^2 \right). \quad (10.55)$$

The damping term $\Gamma_{\mathbf{k}}$ may even result from a microscopic calculation including a thermal bath [19].

Following Lax's prescription we can promote Eqs. (10.53) to global bare-operator equations

$$\begin{aligned} \frac{d}{dt} \hat{P}_{\mathbf{k}} &= -i\tilde{\omega}_{\mathbf{k}} \hat{P}_{\mathbf{k}} + g_{\mathbf{k}} \hat{P}_{\mathbf{k}_i}^{\dagger} \mathcal{P}_{\mathbf{k}_p}^2 + \hat{\mathcal{F}}_{\hat{P}_{\mathbf{k}}} \\ \frac{d}{dt} \hat{P}_{\mathbf{k}_i}^{\dagger} &= i\tilde{\omega}_{\mathbf{k}_i} \hat{P}_{\mathbf{k}_i}^{\dagger} + g_{\mathbf{k}_i}^* \hat{P}_{\mathbf{k}} \mathcal{P}_{\mathbf{k}_p}^2 + \hat{\mathcal{F}}_{\hat{P}_{\mathbf{k}_i}^{\dagger}}. \end{aligned} \quad (10.56)$$

However, in this form it is not a ready-to-use ingredient, indeed its implementation in calculating spectra and/or higher order correlators would be problematic because the noise commutation relations require the solution of the same (at best of an analogous) kinetic problem to be already at hand. This point can be very well explained as soon as one is interested in calculating $\langle \hat{P}_{\mathbf{k}}^{\dagger} \hat{P}_{\mathbf{k}} \rangle$, i.e. the polariton occupation, where the mere calculation is self-explanatory. We shall need

$$2\langle \hat{P}_{\mathbf{k}}^{\dagger} \hat{P}_{\mathbf{k}} \rangle_R = \frac{d}{dt} \langle \hat{P}_{\mathbf{k}}^{\dagger} \hat{P}_{\mathbf{k}} \rangle_R - \left\langle \left\{ \frac{d}{dt} \hat{P}_{\mathbf{k}}^{\dagger} \right\} \hat{P}_{\mathbf{k}} \right\rangle_R - \left\langle \hat{P}_{\mathbf{k}}^{\dagger} \left\{ \frac{d}{dt} \hat{P}_{\mathbf{k}} \right\} \right\rangle_R, \quad (10.57)$$

and the diffusion coefficient for the two operators in reverse order. Thanks to the structure above we can easily see that all coherent contributions cancel out and only the incoherent ones are left. Anyway the important fact for the present purpose is that they are proportional to the polaritonic occupation. In particular the relevant

diffusion coefficients can be written explicitly as

$$2\langle\hat{D}_{P_k^\dagger P_k}\rangle = \sum_{\mathbf{k}'} W_{\mathbf{k},\mathbf{k}'} \langle\hat{P}_{\mathbf{k}'}^\dagger \hat{P}_{\mathbf{k}'}\rangle_{PL}, \quad (10.58)$$

$$2\langle\hat{D}_{P_k P_k^\dagger}\rangle = \sum_{\mathbf{k}'} W_{\mathbf{k},\mathbf{k}'} \left(\langle\hat{P}_{\mathbf{k}'}^\dagger \hat{P}_{\mathbf{k}'}\rangle_{PL} + 1 \right) + \gamma_{\mathbf{k}}^c \quad (10.59)$$

where the time dependence of expectation values is implicit and the label PL indicates that noise is determined only by incoherent contributions i.e. only by the incoherent population dynamics determined by rate equations. $W_{\mathbf{k},\mathbf{k}'}$ and $\gamma_{\mathbf{k}}^c$ are scattering rates arising from the interaction with a thermal bath, e.g. due to phonon-induced scattering, see Ref. [19] for details of the microscopic mechanism they represent. The general solution of eqs. (10.56) in the pump reference frame reads:

$$\begin{aligned} \mathbf{P}(t) &= e^{\int_0^t \mathcal{M}(t') dt'} \mathbf{P}(0) + \int_0^t e^{\int_{t'}^t \mathcal{M}(t'') dt''} \mathcal{K}(t') dt' \\ \mathbf{P}(t) &= \begin{pmatrix} \hat{P}_{\mathbf{k}}(t) \\ \hat{P}_{2\mathbf{k}_p - \mathbf{k}}^\dagger(t) \end{pmatrix}, \mathcal{K} = \begin{pmatrix} \hat{\mathcal{F}}_{\hat{P}_{\mathbf{k}}} \\ \hat{\mathcal{F}}_{\hat{P}_{2\mathbf{k}_p - \mathbf{k}}^\dagger} \end{pmatrix} \\ \mathcal{M} &= \begin{pmatrix} \bar{\omega}_{\mathbf{k}} & \Delta(\mathbf{k}, \tau) \\ \Delta^*(\mathbf{k}, \tau) & \bar{\omega}_{2\mathbf{k}_p - \mathbf{k}}^* \end{pmatrix}, \end{aligned} \quad (10.60)$$

where

$$\begin{aligned} \bar{\omega}_{\mathbf{k}} &= -i\tilde{\omega}_{\mathbf{k}}, \\ \bar{\omega}_{2\mathbf{k}_p - \mathbf{k}} &= -i(\tilde{\omega}_{2\mathbf{k}_p - \mathbf{k}} - 2\tilde{\omega}_{\mathbf{k}_p}), \\ \text{the pump is } \mathcal{P}_{\mathbf{k}_p} &= \mathcal{P}_{\mathbf{k}_p}^o e^{-i\omega_{\mathbf{k}_p}^{(h)} t}, \\ \hat{P}_{2\mathbf{k}_p - \mathbf{k}}^\dagger &= \hat{P}_{2\mathbf{k}_p - \mathbf{k}}^\dagger e^{-i2\omega_{\mathbf{k}_p}^{(h)} t}, \\ \hat{\mathcal{F}}_{\hat{P}_{2\mathbf{k}_p - \mathbf{k}}^\dagger} &= \hat{\mathcal{F}}_{\hat{P}_{2\mathbf{k}_p - \mathbf{k}}^\dagger} e^{-i2\omega_{\mathbf{k}_p}^{(h)} t}, \\ \omega_{\mathbf{k}}^{(h)} &= \omega_{\mathbf{k}} + h_{\mathbf{k}} |\mathcal{P}_{\mathbf{k}_p}|^2, \\ \Delta(\mathbf{k}, \tau) &= g_{\mathbf{k}} \mathcal{P}_{\mathbf{k}_p}^o{}^2. \end{aligned} \quad (10.61)$$

Eq. (10.60) with Eqs. (10.58) and (10.59) provide an easy and general starting point for the calculation of multi-time correlation functions which are key quantities in quantum optics. More explicit expressions for the generated polariton occupations and for the corresponding frequency spectrum including both PL noise and parametric emission can be derived from Eqs. (10.58, 10.59, 10.60) and is reported elsewhere [19]. In the low and intermediate excitation regime, at pump densities where parametric emission starts to appear against the PL background, the main incoherent contribution to the dynamics is the PL the pump produces by itself e.g. by phonon

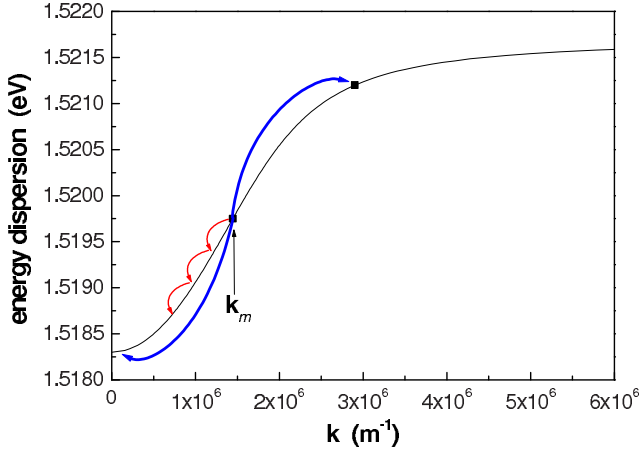


Fig. 10.3 Energy dispersion of the lowest polariton branch for a structure consisting in a 25 nm GaAs/Al_{0.3}Ga_{0.7}As single quantum well placed in the center of a λ cavity with AlAs/Al_{0.15}Ga_{0.85}As Bragg reflectors. The pump at the magic angle and its parametric scattering (blue curve) are schematically depicted. The latter scatters two pump polaritons in a signal-idler couple at $\mathbf{k} = 0$ and $\mathbf{k} = 2\mathbf{k}_m$. The red curve symbolizes incoherent pump scattering at $T = 0$ K, e.g. due to acoustic phonon interaction.

scattering towards other states. The PL arising from incoherent scattering of polariton states populated by parametric processes can be neglected. In this case incoherent dynamics decouples from parametric processes. In particular one can solve the rate equations describing the incoherent dynamics without including parametric processes, and then use the obtained populations to calculate the diffusion coefficients (10.58) and (10.59). We present numerical results taking into account self-stimulation but neglecting the less relevant pump-induced renormalization of polariton energies. We consider a SMC analogous to that of Refs. [13, 43] consisting of a 25 nm GaAs/Al_{0.3}Ga_{0.7}As single quantum well placed in the center of a λ cavity with AlAs/Al_{0.15}Ga_{0.85}As Bragg reflectors. The lower polariton dispersion curve is shown in Fig. 10.3. The calculations are performed at $T = 5$ K and the used cavity linewidth: $\hbar\gamma_c = 0.26$ meV is taken from measured values. The laser pump is modeled as a single Gaussian-shaped pulse of FWHM $\tau = 1$ ps exciting a definite wave vector $\mathbf{k}_p$ and centered at $t = 4$ ps. We pump with co-circularly polarized light exciting polaritons with the same polarization, the laser intensities I are chosen as multiple of I_0 corresponding to a photon flux of $21 \mu\text{m}^{-2}$ per pulse. It has been theoretically shown [35], that it is quite difficult to populate the polaritons in the strong coupling region (i.e. near the bottom of the energy band) by means of phonon-scattering due to a bottleneck effect, similar to that found in the bulk. Let us consider a pump beam resonantly exciting polaritons at about the magic-angle. Relaxation by one-phonon scattering events is effective when the energy difference of the involved polaritons do not exceeds 1 meV. When polariton states within this energy window get populated, they can relax by emitting a phonon to lower energy

levels or can emit radiatively. Owing to the reduced density of states of polaritons and to the increasing of their photon-component at lower energy, radiative emission largely exceeds phonon scattering, hence inhibiting the occupation of the lowest polariton states. Actually this effect is experimentally observed only very partially and under particular circumstances [47]. This is mainly due to other more effective scattering mechanisms [48] usually present in SMCs. For example the presence of free electrons in the system determines an efficient relaxation mechanism. Here we present results obtained including only phonon-scattering. Nevertheless the theoretical framework here developed can be extended to include quite naturally other enriching contributions that enhance non-radiative scattering and specifically relaxation to polaritons at the lowest k -vectors [48]. In order to avoid the resulting unrealistic low non-radiative scattering particularly evident at low excitation densities, we artificially double the acoustic-phonon scattering rates. However, acting this way, we obtain non-radiative relaxation rates that on average agree with experimental values. Figure 10.4 shows the calculated time dependent polariton mode-occupation

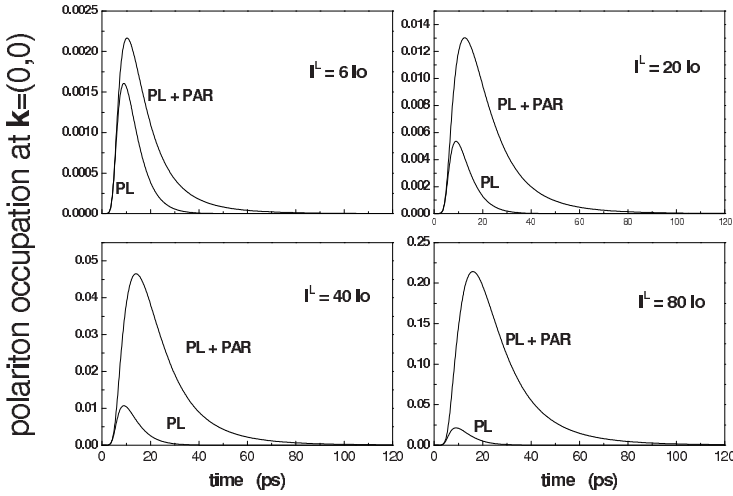


Fig. 10.4 Calculated time dependent polariton mode-occupation at $\mathbf{k} = (0,0)$ obtained at four different pump intensities in comparison with the time dependent pump-induced PL at the same $\mathbf{k}$.

of emitted polaritons at $\mathbf{k} = (0,0)$ $\langle P_0^\dagger P_0 \rangle$, obtained for four different pump intensities in comparison with the time dependent pump-induced PL at the corresponding $\mathbf{k}$. The pump beam is sent at the magic angle [49] ($k_m \simeq 1.44 \cdot 10^6 \text{ m}^{-1}$) which is close to the inflection point of the energy dispersion curve and is resonant with the polariton state at k_m . The magic angle is defined as the pump value needed for the eight-shaped curve of the resonant signal-idler pairs to intersect the minimum of the polariton dispersion curve at $\mathbf{k} = 0$. It is worth noting that the displayed results have no arbitrary units. We address realistic input excitations and we obtain quantitative outputs, indeed in Figs. 10.4 we show the calculated polariton occupa-

tion, i.e. the number of polaritons per mode. In our calculations no fitting parameter is needed, nor exploited (apart from the doubling of the phonon scattering rates). Moreover our results are in good agreement with the experimental results of Ref. [43] the pump intensity at which parametric scattering, superseding the pump-PL, becomes visible. We notice that already at moderate pump excitation intensities the parametric contribution dominates. It represents a clear evidence that we may devise future practical experiments exploiting such a window where the detrimental pump-induced PL contribution is very low while at the same time we are in a regime with quite small amount of emitted polaritons per mode. Indeed, for photon-counting coincidence detections to become good experimental means of investigation, we need a situation where accidental detector's clicks are fairly absent and where the probability of states with more than one photon is low. We now focus our attention on

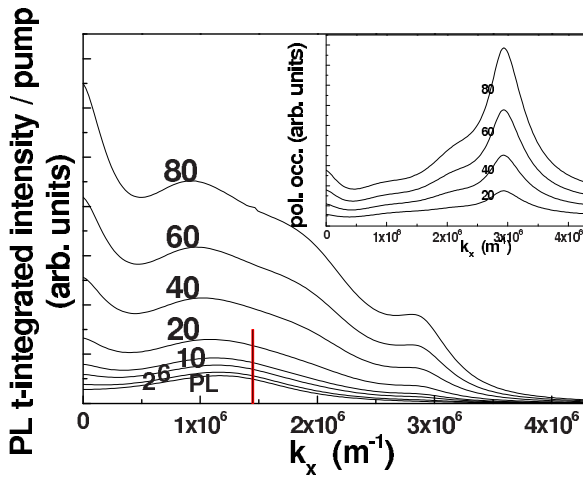


Fig. 10.5 Time-integrated outgoing photon emission intensity vs k_x at different normalised pump intensities: 2,6,10,20,40,60,80.. The pump is set at $\mathbf{k}_p = (k_m, 0)$. It is clear the evidence of the build-up of the parametric emission taking over the pump-induced PL once the seed beam becomes higher than the threshold around $I^L = 20I_0$. Moreover the parametric process removes the phonon bottleneck in the region closed to $\mathbf{k} = 0$. The specific signal-idler parametric scattering with the signal in $k_x = 0$ and the idler in $k_x = 2k_m$ is favoured and at higher pump intensities dominates the light emission. The pump is set at the red line. The polariton idler occupations for some pump values are depicted in the inset. Although polariton occupation at $k_x = 2k_m$ is so high, its photonic component is very small resulting in a very weak outgoing light beam.

the positive part of the $k_y = 0$ section at different pump powers. In Fig. 10.5 we observe the clear evidence of the build-up of the parametric emission taking over the pump-induced PL once the seed beam has become enough intense, in particular we can set a threshold around $I = 10I_0$ ($I^L = 20I_0$). As expected, the parametric process with the pump set at the magic angle enhances the specific signal-idler pair with the signal in $k_x = 0$ and the idler in $k_x = 2k_m$. We can clearly see from the figure that at pump intensities higher than the threshold the idler peak becomes more and more

visible for increasing power in agreement of what shown in Ref. [49]. However, at so high k_x values the photon component is very small and even if the polariton idler occupation is quite high (as the inset of Fig. 10.5 shows), the outgoing idler light is so weak to cause some difficulties in experiments [43]. Moreover we can notice that the parametric process removes the phonon bottleneck in the region close to $\mathbf{k} = 0$. An analogous situation occurs also in Ref. [47], though with a different SMC, where the bottleneck removal in $\mathbf{k} = 0$ due to the parametric emission can be clearly seen.

10.7 Quantum complementarity of cavity polaritons

When two physical observables are complementary, the precise knowledge of one of them makes the other unpredictable. The most known manifestation of this principle is the ability of quantum-mechanical entities to behave as particles or waves under different experimental conditions. For example, in the famous double-slit experiment, a single electron can apparently pass through both apertures simultaneously, forming an interference pattern. But if a “which-way” detector is employed to determine the particle’s path, the particle-like behaviour takes over and an interference pattern is no longer observed. Atomic interferometry experiments have brought evidence that the loss of interference is not necessarily the consequence of the back action of a measurement process [50]. Quantum complementarity is rather an inherent property of a system, enforced by quantum correlations. The link between quantum correlations, quantum nonlocality and Bohr’s complementarity principle was established in a series of “which-way” experiments using quantum-correlated photon pairs emitted via parametric down-conversion [51, 52, 53, 54]. These experiments unequivocally showed that the quantum-mechanical correlation between two different light modes is able to store the “which-way” information, thereby affecting the interference pattern. These results at the same time help a fundamental understanding of the features of pair quantum states and constitute an elegant demonstration of nonclassical correlations between the photons emitted in parametric down conversion.

Here we report on the theoretical and experimental investigation of quantum complementarity in semiconductor microcavities. Experimental results provide evidence for quantum correlations of emitted polariton pairs. Excitons exhibit strong nonlinearities due to saturation effects (phase space filling) and to Coulomb interaction. The latter enforces exciton-exciton scattering and determines many-body correlations including biexciton effects. Polariton-polariton interactions are determined by their exciton content. Owing to these interactions, pump polaritons generated by a resonant optical excitation can scatter into pairs of polaritons (the signal and the idler modes) according to total momentum and energy conservation criteria [16, 29, 36].

Given a pump wave vector $\mathbf{k}_p$, the set of possible parametric processes that satisfy total energy and momentum conservation is represented by an “eight”-shaped curve in $\mathbf{k}$ -space [36], as displayed in Fig. 10.6. On this curve, a pair of signal-

and idler-modes is defined by the intersection with a straight line passing through $\mathbf{k}_p$. As a convention, we designate as “idler” the modes with $k > k_p$. The experimental scheme that we devise is based on two energy-degenerate pump modes having momentum $\mathbf{k}_{p1}$ and $\mathbf{k}_{p2}$ respectively. In this configuration together with the two customary eight-shape curves involving a single pump, mixed-pump parametric processes are allowed depicting the “peanut”-shaped curve in $\mathbf{k}$ -space in Fig. 10.6. Let us consider the two mutually coherent pump polariton fields as of equal energy and amplitude but with different in-plane wave vector $\mathcal{P}_{\mathbf{k}_{p2}} = \mathcal{P}_{\mathbf{k}_{p1}} e^{i\phi}$. Within this scheme, pairs of parametric processes sharing the signal mode are allowed [13]. Such a pair of processes involves two idler modes $i1$ and $i2$ and one common signal mode s . For this setup the equation of motion for the emitted polariton modes reads

$$\begin{aligned} \frac{d}{dt} \hat{P}_{\mathbf{k}_s} &= -i\tilde{\omega}_{\mathbf{k}} \hat{P}_{\mathbf{k}_s} + g \left(\hat{P}_{\mathbf{k}_{i1}}^\dagger \mathcal{P}_{\mathbf{k}_{p1}}^2 + \hat{P}_{\mathbf{k}_{i2}}^\dagger \mathcal{P}_{\mathbf{k}_{p2}}^2 \right) + \hat{\mathcal{F}}_{\mathbf{k}_s} \\ \frac{d}{dt} \hat{P}_{\mathbf{k}_{i1}}^\dagger &= i\tilde{\omega}_{\mathbf{k}_{i1}} \hat{P}_{\mathbf{k}_{i1}}^\dagger + g \hat{P}_{\mathbf{k}_s} \mathcal{P}_{\mathbf{k}_{p1}}^{*2} + \hat{\mathcal{F}}_{\mathbf{k}_{i1}}^\dagger \\ \frac{d}{dt} \hat{P}_{\mathbf{k}_{i2}}^\dagger &= i\tilde{\omega}_{\mathbf{k}_{i2}} \hat{P}_{\mathbf{k}_{i2}}^\dagger + g \hat{P}_{\mathbf{k}_s} \mathcal{P}_{\mathbf{k}_{p2}}^{*2} + \hat{\mathcal{F}}_{\mathbf{k}_{i2}}^\dagger, \end{aligned} \quad (10.62)$$

The nonlinear couplings g entering the three equations results to be equal, so we dropped its $\mathbf{k}$ -dependence.

For the sake of presentation, in the limit of low excitation intensity we can construct an effective *bosonic* Hamiltonian able to give, once applied by customary quantum rules to the wave function, the schematic path of signal-idler pairs by parametric scattering. Then, the lowest polariton-number state would be cast in the form

$$|\Psi\rangle = \frac{1}{\sqrt{2}} |1\rangle_s (|1\rangle_{i1} |0\rangle_{i2} + \exp(-2i\phi) |0\rangle_{i1} |1\rangle_{i2}), \quad (10.63)$$

where the two entangled idler states are sharing the same signal s . The polariton density at the signal mode $\langle \Psi | \hat{P}_{\mathbf{k}_s}^\dagger \hat{P}_{\mathbf{k}_s} | \Psi \rangle$ results to be clearly independent of ϕ . Interference is absent due to the orthogonality of the two idler states whose sum is enclosed in parentheses. This sum stores the “which-way” information, each term representing one possible idler path in the process. Hence the pair-correlation between signal and idler polaritons and the entanglement of the two idler paths are the reason for the absence of interference, as implied by expression (10.63). Interference is also absent from either idler-polariton density. A different result is found when looking at the mutual coherence of the two idler beams which is observable in the sum of the two idler polariton fields. The resulting particle density is $\langle \Psi | (\hat{P}_{\mathbf{k}_{i1}}^\dagger + \hat{P}_{\mathbf{k}_{i2}}^\dagger) (\hat{P}_{\mathbf{k}_{i1}} + \hat{P}_{\mathbf{k}_{i2}}) | \Psi \rangle = 2 \langle \Psi | \hat{P}_{\mathbf{k}_{i1}}^\dagger \hat{P}_{\mathbf{k}_{i1}} | \Psi \rangle [1 + \cos(2\phi)]$. Interference now appears owing to the cross term $\langle \Psi | \hat{P}_{\mathbf{k}_{i1}}^\dagger \hat{P}_{\mathbf{k}_{i2}} | \Psi \rangle$.

In this case interference occurs because the two idler modes are pair-correlated with the same signal mode. Even by means of a signal-idler coincidence measurement, no “which-way” information could be retrieved. Hence the presence or absence of interference is a direct consequence of the complementarity principle en-

forced by the pair-correlation between signal and idler modes. A similar analysis on the more general many-polariton state leads to the same conclusions [55].

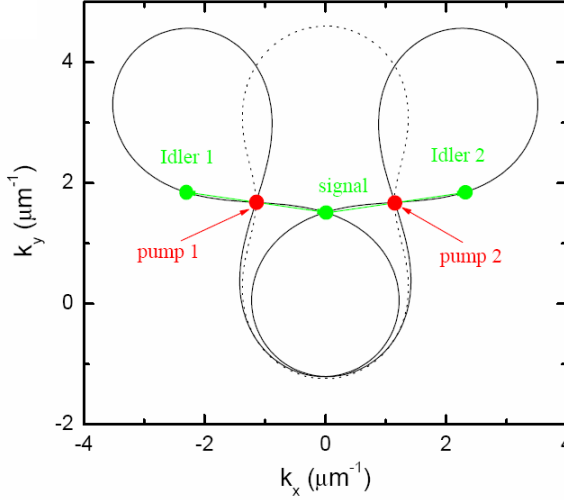


Fig. 10.6 $\mathbf{k}$ -space plot of the final states fulfilling energy and momentum conservation in a two-pump parametric process. The two “eight”-shaped full curves represent the single-pump processes determined by the conditions $2\mathbf{k}_{pj} = \mathbf{k}_s + \mathbf{k}_i$ and $2E_{k_{pj}} = E_{k_s} + E_{k_i}$, with $j = 1, 2$. The dotted line describes the mixed-pump process defined by $\mathbf{k}_{p1} + \mathbf{k}_{p2} = \mathbf{k}_s + \mathbf{k}_i$ and $E_{k_{p1}} + E_{k_{p2}} = E_{k_s} + E_{k_i}$. Two parametric processes sharing $\mathbf{k}_s$, giving rise to mutual idler coherence are indicated in green.

The coherence properties of the polaritons are stored in the emitted photons, thus making it possible to carry out the devised “which-way” measurement within a standard optical spectroscopy setup. Our task is therefore to sum the two emitted idler fields and detect their mutual interference as a function of the relative pump phase. This is accomplished by detecting two superimposed $\mathbf{k}$ -resolved images of the polariton emission, one of which is preliminarily mirrored around the k_y axis. The investigated device [13] consists of a 25 nm GaAs/Al_{0.3}Ga_{0.7}As single quantum well placed in the center of a λ -cavity with AlAs/Al_{0.15}Ga_{0.85}As Bragg reflectors. The use of a wide GaAs quantum well leads to a negligible inhomogeneous broadening of the quantum well exciton due to the small effect of interface roughness, which allows to have well-defined polariton modes also at large in-plane momenta. Blocking one of the detection interferometer arms, the intensity $I_{\mathbf{k}} = |\mathcal{E}_{\mathbf{k}}|^2$ is measured (proportional to the polariton density $N_{\mathbf{k}} = \langle \Psi | \hat{P}_{\mathbf{k}}^\dagger \hat{P}_{\mathbf{k}} | \Psi \rangle$).

The nonlinear couplings entering the three equations result to be equal, so we dropped their $\mathbf{k}$ -dependence. It is possible to define new idler operators as follows:

$$\hat{P}_{1,2} = \hat{P}_{\mathbf{k}_{i1}} \pm e^{-2i\phi} \hat{P}_{\mathbf{k}_{i2}}.$$

Correspondingly we define new noise operators $\hat{\mathcal{F}}_{1,2}$. From Eq. (10.62) we obtain

$$\begin{aligned}\frac{d}{dt}\hat{P}_{\mathbf{k}_s} &= -i\tilde{\omega}_{\mathbf{k}}\hat{P}_{\mathbf{k}_s} + \sqrt{2}g\hat{P}_1^\dagger \mathcal{P}_{\mathbf{k}_{p1}}^2 + \hat{\mathcal{F}}_{\mathbf{k}_s} \\ \frac{d}{dt}\hat{P}_1^\dagger &= i\tilde{\omega}_1\hat{P}_1^\dagger + \sqrt{2}g\hat{P}_{\mathbf{k}_s} \mathcal{P}_{\mathbf{k}_{p1}}^{*2} + \hat{\mathcal{F}}_1^\dagger \\ \frac{d}{dt}\hat{P}_2^\dagger &= i\tilde{\omega}_2\hat{P}_2^\dagger + \hat{\mathcal{F}}_2^\dagger,\end{aligned}\tag{10.64}$$

where $\tilde{\omega}_{1,2} = \tilde{\omega}_{\mathbf{k}_{i1}} = \tilde{\omega}_{\mathbf{k}_{i2}}$. In terms of the new operators the parametric process with one signal mode coupled to two idler modes can be viewed as an ordinary parametric process involving $\hat{P}_{\mathbf{k}_s}$ and $\hat{P}_1$ plus one decoupled mode $\hat{P}_2$. There is no parametric process driving P_2 , this mode can thus be populated only by photoluminescence.

The absence of interference despite the resulting population arises from two coherent pumps can be interpreted on the basis of the quantum complementarity principle. In principle it would be possible to know whether a detected signal polariton comes from pump 1 or 2, looking at coincidences with the idler beam. According to the principle this possible which-way information destroys interference. If the two idler beams are superimposed before detection, the detected light field is proportional to

$$I = \langle (\hat{P}_{\mathbf{k}_{p1}}^\dagger + \hat{P}_{\mathbf{k}_{p2}}^\dagger)(\hat{P}_{\mathbf{k}_{p1}} + \hat{P}_{\mathbf{k}_{p2}}) \rangle / 2.\tag{10.65}$$

In terms of the new operators we obtain

$$I = [N_1(1 + \cos 2\phi) + N_2(1 - \cos 2\phi)]/2,$$

($N_{1/2} = \langle \Psi | \hat{P}_{1/2}^\dagger \hat{P}_{1/2} | \Psi \rangle$) showing interference because the two idler modes are pair correlated with the same signal mode, and thus even by a signal-idler coincidence measurement, no which-way information could be retrieved.

The resulting visibility, defined as

$$\mathcal{V} \equiv \frac{I_{\max} - I_{\min}}{I_{\max} + I_{\min}}\tag{10.66}$$

is $\mathcal{V} = (N_1 - N_2)/(N_1 + N_2)$. In the absence of incoherent processes $N_2 = 0$ and $\mathcal{V} = 1$. Incoherent effects thus lower the visibility. A classical model of parametric emission can be obtained from Eqs. (10.64) replacing quantum operators with $\mathbb{C}$ -numbers. In this case the quantum Langevin forces becomes classical stochastic noise terms. While the quantum parametric process is initiated by vacuum fluctuations, the classical version describes (in the absence of a coherent signal or idler input) essentially the parametric amplification of noise. Figure 10.7 displays the calculated time-resolved idler polariton number per mode (dashed) originating from parametric emission, and the calculated interference visibility. The pump is set so that the peak idler-polariton occupation number per mode is $N_{\mathbf{k}_{i1}} = N_{\mathbf{k}_{i2}} \simeq 2$. The figure also reports the corresponding classical calculation largely underestimating the measured visibility [13]. This calculation thus confirms the quantum nature of the measured interference. We observe that the present calculation includes a micro-

scopic description of PL giving rise to a realistic description of its time dependence (see Fig. 10.4), while in Ref. [13] PL has been included as a constant phenomenological noise.

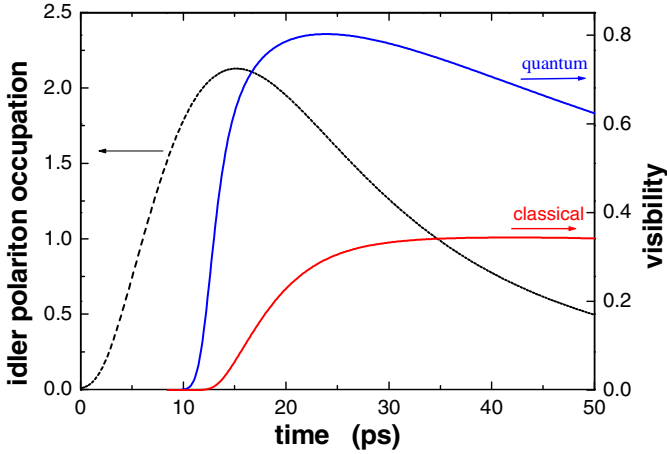


Fig. 10.7 Calculated time-resolved idler polariton number per mode (dashed) originating from parametric emission; quantum and classical interference visibility when combining two idler beams sharing a common signal.

As our experiment uses pulsed excitation, the idler interference has a distinct dynamics during the buildup and the decay of the parametric scattering [43]. In Fig. 10.7 we show the measured interference visibility as a function of time, together with the idler intensity. The visibility reaches 0.8 soon after the pump pulse, much earlier than the maximum in the idler-polariton number. Such a high visibility in absence of spectral filters is quite surprising and shows that SMCs are very promising for studying quantum optical correlations in the presence of real electronic excitations. A coincidence detection of signal-idler pairs could directly assess the quantum origin of the interference [52], but was not available for the present work. We therefore need to consider that interference could be accounted for by a classical parametric model including random-noise driving terms [56], for which the visibility is significant only in the self-stimulated regime. The present measurement was performed at the onset of the self-stimulated regime, corresponding to an excitation intensity of $40I_0$ according to Ref. [43]. In this regime, the estimated peak idler-polariton occupation number per mode is $N_k \sim 2$. In order to assess the nature of the measured interference, we thus have to compare it with the quantum and the classical predictions. The comparison of the two results (see Fig. 10.7) with the measurement in Ref. [13] confirms the quantum origin of the measured interference.

10.8 Coherent Trapping

In this section we provide an application of the two-pump scheme to polariton amplification [57]. The results in this section (in contrast to those in the previous one) can be described within a semiclassical model. We notice that in principle the role of signal and idler polaritons can be reversed, so that we can stimulate the scattering process sending a weak resonant light pulse in the idler direction, thus observing amplification of idler polaritons as well as a strong emission at normal incidence.

Owing to the axial symmetry of the polariton energy dispersion we may think of many different parametric processes satisfying energy and momentum conservation. In particular the cavity can be pumped by different beams all at the same *magic* angle θ_p and with the same energy but with different azimuthal angle ϕ . In this case the in-plane pump momenta $\mathbf{k}_j$ have all the same modulus $k = \omega \sin \theta_p / c$ but different directions $\mathbf{k}_j = k(\cos \phi_j, \sin \phi_j)$. These different parametric processes give rise each to a corresponding distinct idler beam with momentum $2\mathbf{k}_j$ but only one signal beam in the direction normal to the SMC. Thus we may entangle many distinct parametric processes realizing a sort of parametric trap where many different processes are forced to produce scattering towards a common polariton state. So the emission at normal incidence is the result of many eventually interfering parametric processes.

We can seed the SMC with two pump laser pulses at $\mathbf{k}_1$ and $\mathbf{k}_2$ (1,2 pump modes), on the lower polariton dispersion. Then two laser pulses respectively at $2\mathbf{k}_{1,2}$ (1,2 idler modes) are sent in the SMC. If the pump laser pulses excite the polariton dispersion at the magic angle (hence $|\mathbf{k}_1| = |\mathbf{k}_2| = k$), the two idler beams stimulate two independent scattering process in which two (1 or 2) pump polaritons are scattered in the 1 or 2 idler mode and in a single (*degenerate*) signal mode at $\mathbf{k} = 0$ with the prescribed energy conservation $2\omega_{\mathbf{k}_{1,2}} = \omega_{2\mathbf{k}_{1,2}} + \omega_0$. Each of the processes above described is analogous to the parametric amplification process obtained in several experiments with the difference that in this situation the process is stimulated sending a weak light pulse in the idler directions [determined by $\phi_{1,2}$, $\arcsin(2kc/\omega_{2k})$] instead of the signal one. We observe that this particular configuration determines two different contributions to the signal amplification due to the two distinct amplification processes. Our aim is to show that acting on the delay and phase of the ultrafast idler laser pulses it is possible to obtain the coherent control of the signal emission. From the Heisenberg equations of motion for the quantum polariton operators performing the rotating wave approximation and the taking the expectation values of the quantum operators involved, we obtain

$$i\hbar\dot{p}_0 = -i\Gamma_0 p_0 + g(p_{2\mathbf{k}_1}^* p_{\mathbf{k}_1}^2 + p_{2\mathbf{k}_2}^* p_{\mathbf{k}_2}^2) + iC_0 \sqrt{\gamma} E_0^{in}, \quad (10.67a)$$

$$i\hbar\dot{p}_{2\mathbf{k}_1} = -i\Gamma_{2k} p_{2\mathbf{k}_1} + g\hat{p}_0^* p_{\mathbf{k}_1}^2 + iC_{2k} \sqrt{\gamma} E_{2\mathbf{k}_1}^{in}, \quad (10.67b)$$

$$i\hbar\dot{p}_{2\mathbf{k}_2} = -i\Gamma_{2k} p_{2\mathbf{k}_2} + g\hat{p}_0^* p_{\mathbf{k}_2}^2 + iC_{2k} \sqrt{\gamma} E_{2\mathbf{k}_2}^{in}; \quad (10.67c)$$

where g is the effective interaction parameter, $p_{j,\mathbf{k}} = \langle p_{j,\mathbf{k}} \rangle$, Γ_i , and C_i are respectively the dephasing, and the photon Hopfield coefficient relative to the i -th

polariton branch; E_i^{in} describes the input light pulse exciting the i -th polariton branch; $\sqrt{\gamma}$ gives the beam fraction passing through the cavity with equal mirrors. In this approach the two pump beams are considered classical, having amplitudes $p_{\mathbf{k}_1} = e^{-i\omega t} \tilde{p}_{\mathbf{k}_1}$ and $p_{\mathbf{k}_2} = \sqrt{\alpha} e^{-i\omega t + i\phi} \tilde{p}_{\mathbf{k}_1}$, where α is a real constant. They are mutually coherent and proportional to the same classical amplitude, with a mutual difference of phase ϕ that can be experimentally achieved by a delay line. We assume α positive and real.

In order to obtain analytical simple expressions, we can assume for the input idler pulses an ultrafast deltalike behaviour:

$$\begin{cases} E_{2\mathbf{k}_1}^{in} = \varepsilon \delta(t - t_0), \\ E_{2\mathbf{k}_2}^{in} = \varepsilon e^{-i\psi} \delta(t - t_0 - \delta), \end{cases} \quad (10.68)$$

where ψ and δ are the relative phase and time delay between the two input idler pulses respectively. In the case with $\alpha = 1$ and in the absence of delay between the two idler pulses $\delta = 0$, we obtain:

$$\begin{aligned} \tilde{p}_0 &\propto e^{-\Gamma(t-t_0)} \varepsilon \Theta(t-t_0) \\ &\quad \left\{ 1 + e^{-i(\psi-2\phi)} \right\} \sinh(\bar{\Delta}(t, t_0)), \end{aligned} \quad (10.69a)$$

$$\begin{aligned} \tilde{p}_{2\mathbf{k}_1} &\propto e^{-\Gamma(t-t_0)} \varepsilon \Theta(t-t_0) \left\{ (\cosh(\bar{\Delta}(t, t_0)) + 1) \right. \\ &\quad \left. + e^{-i(\psi-2\phi)} (\cosh(\bar{\Delta}(t, t_0)) - 1) \right\} \end{aligned} \quad (10.69b)$$

$$\begin{aligned} \tilde{p}_{2\mathbf{k}_2} &\propto e^{-\Gamma(t-t_0)} \varepsilon \Theta(t-t_0) \left\{ (\cosh(\bar{\Delta}(t, t_0)) - 1) \right. \\ &\quad \left. + e^{-i(\psi-2\phi)} (\cosh(\bar{\Delta}(t, t_0)) + 1) \right\}. \end{aligned} \quad (10.69c)$$

We have defined $\bar{\Delta}(t_1, t_2) = -i\tilde{g} \int_{t_2}^{t_1} (|\tilde{p}_{\mathbf{k}_1}|^2(t')) dt'$; $\tilde{g} = ig\sqrt{1+\alpha^2}$. We are also assuming that the pump polariton amplitudes $p_{\mathbf{k}}$ are not affected by the parametric process.

Eq.s (10.69) show the possibility to obtain a complete coherent control at zero delay ($\delta = 0$) of emission at normal incidence acting on the phase differences $\psi - 2\phi$. Choosing e.g. the two pump beams with the same phase $\phi = 0$, a complete stop of the amplification and emission can be obtained sending two idler beams at zero relative delay with a phase difference $\psi = \pi$. It is possible to show that almost complete coherent control can be achieved sending the two idler pulses with a small relative delay compared to the decay rate of polariton waves.

We can also observe that it is possible, seeding the SMC with two pump beams with opposite relative phase $\phi = n\pi$, to achieve destructive coherent control even if the phase of the two idler pulses is the same ($\psi = 0$). This configuration realizes the effect of parametric polariton trapping. This coherent trapping occurs due to destructive quantum interference between the two parametric processes contributing to the signal emission in the direction normal to the SMC.

10.9 Spin-entangled cavity polaritons

Cavity polaritons can be optically excited at the desired energy and momentum by tuning the incidence angle and the frequency of the exciting laser. Let us analyse again the spontaneous parametric emission in SMCs. We neglect the effects of energy splitting between TE and TM polariton modes. This effect is relevant only at quite large angles and can be prevented by using a scheme with two energy degenerate pumps ($\omega_{\mathbf{k}_p}$) at quite small angles. In this case parametric processes from only one pump are not allowed by energy and momentum conservations, but mixed processes involving both pumps are allowed. We also neglect bound biexciton effects. This last effect is expected to be negligible if the energy of pump polaritons is lower than half the biexciton energy. This can be easily ensured setting the SMC at negative exciton-cavity detuning. With the above assumptions, the polariton parametric scattering preserves the spin orientation, e.g. clockwise polarised pump beams give rise to two clockwise co-polarised signal and idler beams with total energy and momentum conservation, as observed experimentally at low excitation density even under less stringent conditions.

In the following we address calculations within a single pump, generalization to a two-pump scheme is straightforward. Let us consider a **linearly polarized** (along the direction $\tilde{e}_p$) pump laser beam of given in-plane momentum $\mathbf{k}_p$ and energy $\omega_{\mathbf{k}_p}$. The interaction Hamiltonian describing the process for a signal-idler pair is given by

$$\hat{H} = \frac{gP_p^2}{2} [\hat{P}_{s+}^\dagger \hat{P}_{i+}^\dagger + \hat{P}_{s-}^\dagger \hat{P}_{i-}^\dagger] + H.c., \quad (10.70)$$

where P_p^2 represent the classical amplitude of pump polaritons (we also assume the undepleted pump approximation which is a good approximation in the low excitation density regime), $\hat{P}_s, \hat{P}_i$ are the operators describing respectively the signal and idler polariton pairs produced by the scattering process with momenta $\mathbf{k}_s, \mathbf{k}_i$. The Hamiltonian describes two phase coherent twin-beam sources corresponding to the pairs of modes $\hat{P}_{s+}^\dagger, \hat{P}_{i+}^\dagger$ and $\hat{P}_{s-}^\dagger, \hat{P}_{i-}^\dagger$. The two different sources arise from the decomposition of the linear pump polarization $\hat{e}_p$ in the two independent left and right circular components $\hat{e}_-, \hat{e}_+$:

$$\tilde{e}_p = \frac{1}{\sqrt{2}}(\tilde{e}_- + \tilde{e}_+). \quad (10.71)$$

Solving the corresponding Heisenberg-Langevin equations in the low excitation density regime, one finds that the signal and idler emission results to be completely unpolarized. On the contrary, a calculation of signal-idler coincidence displays important polarization effects. This is due to the fact that $+$ and $-$ polarized polaritons are emitted with the same intensity, but polaritons are emitted in pairs that are co-circularly polarized: they are spin-entangled. Neglecting losses and noise, in the limit of low excitation intensity, Hamiltonian (10.70) gives rise to the following spin-entangled polariton quantum state:

$$|\Psi\rangle = \frac{1}{\sqrt{2}} (|+\rangle_s |+\rangle_i + |-\rangle_s |-\rangle_i). \quad (10.72)$$

The present proposal is very tempting as it would allow to obtain optically mixed matter-wave entangled states, i.e. polaritons. However, two important problems need to be addressed in a more microscopic (and sophisticated way): how properly limit the counter-circular polarization channel responsible for spin-mixing, and the time-dependent noise background. Third order nonlinear optical processes involve polaritons with the same circular polarization (co-circular channel) and other pairs of polaritons with opposite circular polarization (counter-circular channel) due to the presence of both bound biexciton states and four-particle scattering states of zero angular momentum ($J = 0$) [11]. A linearly polarized single pump excitation cannot avoid the simultaneous presence of the co-circular scattering channel which can lower polarization entanglement simply because the matrix element of the ($J = 0$) counter-circular scattering channel is smaller (about 1/3) than that for the co-polarized scattering channel, but certainly not negligible [16, 32, 46, 58]. As a consequence, the superposition of co-circular and counter-circular scattering creates a superposition of singlet and triplet states which, in general, has an entanglement degree less than that of its single constituents.

However, in a real system, environment always act as an uncontrollable and unavoidable continuous perturbation producing decoherence and noise. The latter represents a fundamental limitation, as it tends to lower the degree of non-classical correlation or even completely wash it out [13, 59].

The way we can face these important limitations will be the main subject of the next section where we shall present a microscopic analysis of coherent and incoherent processes (on an equal footing) able to lead us to devise the proper set-up able to measure bipartite polariton entanglement within a realistic experiment.

10.10 Emergence of entanglement out of a noisy environment: The case of microcavity polaritons

The concept of *entanglement* has played a crucial role in the development of quantum physics. It can be described as the correlation between distinct subsystems which cannot be reproduced by any classical theory (i.e. *quantum correlation*). It has gained renewed interest mainly because of the crucial role that such concept plays in quantum information/computation (QIC) [60], as a precious resource enabling to perform tasks that are either impossible or very inefficient in the classical realm [61]. Scalable solid-state devices will make use of local electronic states to store quantum correlations [62]. Polaritons [63] on the other hand, as hybrid states of electronic excitations and light, are the most promising solution for generation and control of quantum correlations over long range [64]. In particular, thanks to the Coulomb interaction acting on the electronic part of the polariton state, resonantly generated pump polaritons scatter into pairs of *signal* and *idler* polaritons, in

a way that fulfills total energy and momentum conservation. The generated polariton pairs can be in an entangled state [65, 42]. In this case the outcome of a polariton parametric scattering is an *entangled state of a hybrid quasiparticle, half light and half electronic excitation, of the semiconductor* – the polariton pair – contrarily to parametric down-conversion in a nonlinear crystal [65], where only the outgoing photons are entangled. Here, the emitted photons serve merely as a probe of the internal degree of entanglement. Thanks to their photon component, polaritons can sustain quantum correlations over mesoscopic distances inside the semiconductor. This is why they bear a unique potential as a controllable embedded mechanism to generate quantum information in a device and transfer it to localized qubits (e.g. spin qubits) over distances of microns [64]. In order to address entanglement in quantum systems [12, 66], the preferred experimental situation is the few-particle regime in which the emitted particles can be detected individually [67, 68]. In a real system, environment always act as an uncontrollable and unavoidable continuous perturbation producing decoherence and noise. Even if polariton experiments are performed at temperature of few Kelvin [13], polaritons created resonantly by the pump can scatter, by emission or absorption of acoustic phonons, into other states, acquiring random phase relations. These polaritons form an incoherent background (i.e. noise), responsible of *pump-induced* photoluminescence (PL), which competes with coherent photoemission generated by parametric scattering, as evidenced by experiments [43]. As a consequence, noise represents a fundamental limitation, as it tends to lower the degree of non-classical correlation or even completely wash it out [13, 59]. Hence understanding the impact of noise on quantum correlations in semiconductor devices, where the electronic system cannot be easily isolated from its environment, is crucial.

In this section, we present a microscopic study of the influence of time-dependent noise on the polarization entanglement of polaritons generated in parametric PL. Our treatment accounts for realistic features such as detectors noise background, detection windows, dark-counting etc., needed [19] in order to seek and limit all the unwanted detrimental contributions. We show how a tomographic reconstruction [69], based on two-times correlation functions, can provide a quantitative assessment of the level of entanglement produced under realistic experimental conditions. In particular, we give a ready-to-use realistic experimental configuration able to measure the Entanglement of Formation (EOF) [70, 71], out of a dominant time-dependent noise background, without any need for post-processing [66].

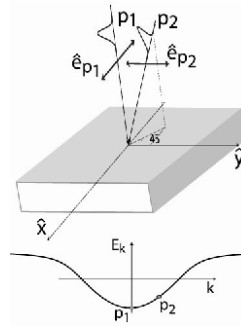
10.10.1 Coherent and incoherent polariton dynamics

Third order nonlinear optical processes in quantum well excitons (with spin $\sigma = \pm 1$) can be described in terms of two distinct scattering channels: one involving only excitons (polaritons) with the same circular polarization (co-circular channel); and the other (counter-circular channel) due to the presence of both bound biexciton states and four-particle scattering states of zero angular momentum ($J = 0$) [11].

Bound biexciton-based entanglement generation schemes [12, 24, 72], producing entangled polaritons with opposite spin, need specific tunings for efficient generation, and are expected to carry additional decoherence and noise due to scattering of biexcitons. Moreover linearly polarized single pump excitation cannot avoid the additional presence of the co-circular scattering channel which can lower polarization entanglement. The experimental set-up that we will choose to calculate the emergence of polariton spin-entanglement is a two-pump scheme under pulsed excitation, involving the lower polariton branch only. The pumps (p_1 and p_2) are chosen with incidence angles below the *magic angle* [43] so that single-pump parametric scattering is negligible. In this setup, mixed-pump processes (signal at in-plane wave vector $\mathbf{k}$, idler at $\mathbf{k}_i = \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}$) are allowed. We choose $\mathbf{k}_1 = (0., 0.)$, and $\mathbf{k}_2 = (0.9, 0.9)\mu\text{m}^{-1}$. As signal-idler pair, we choose to study the two energy-degenerate modes at $\mathbf{k} \simeq (k_{1x}, k_{2y})$ and $\mathbf{k}_i \simeq (k_{2x}, k_{1y})$, as shown in Fig. 10.8 and Fig. 10.9. Of course a number of different two-pump schemes can also conveniently be adopted. For instance, from an experimental viewpoint, a two energy-degenerate pumps setting can be more valuable. Then possible choices for signal-idler pairs within the circle of available parametrically-generated final states (see Fig. 10.9) would suffer only a slightly unbalance being anyway close to the origin of the polariton dispersion curve. For all the numerical simulations we will consider the sample investigated in Ref. [43]. In particular, we shall employ two pump beams linearly cross polarized (then the angle θ will refer to the polarization of one of the two beams, see Fig. (10.8)). This configuration is such that the counter-circular scattering channel (both bound biexciton and scattering states) is suppressed owing to destructive interference, while co-circular polarized signal-idler beams are generated. In the absence of the noisy environment, polariton pairs would be cast in the pure triplet entangled state $|\psi_{\parallel}\rangle = |+, +\rangle - \exp(i4\theta)|-, -\rangle$.

The advantages of this configuration are manifold. First, processes detrimental for entanglement as the excitation induced dephasing results to be largely suppressed [16, 32, 46, 58]. Spurious coherent processes, e.g. Resonant Rayleigh Scattering [73], are well separated in $\mathbf{k}$ -space from the signal and idler modes. In addition signal and idler close to the origin in $\mathbf{k}$ -space make negligible the longitudinal-transverse splitting of polaritons [15] (relevant at quite high in-plane wave vectors).

Fig. 10.8 sketch of the proposed excitation geometry and of the lower polariton branch. The Gaussian pumps are linearly cross polarized with zero time delay. The specific polarization configuration with $\theta = 0$, where θ is the angle between $\hat{e}_{p1}$ and the x -axis on the $x-y$ plane, is depicted.



Following Ref. [69] the tomographic reconstruction of the two-polariton density matrix is equivalent, in the $\sigma = \{+, -\}$ polarization basis, to the two-time coincidence

$$\rho_{\sigma\bar{\sigma},\sigma'\bar{\sigma}'} = \frac{1}{\mathcal{N}} \int_{T_d} dt_1 \int_{T_d} dt_2 \langle \hat{P}_{\mathbf{k}\sigma}^\dagger(t_1) \hat{P}_{\mathbf{k}_i\bar{\sigma}}^\dagger(t_2) \hat{P}_{\mathbf{k}_i\bar{\sigma}'}(t_2) \hat{P}_{\mathbf{k}\sigma'}(t_1) \rangle, \quad (10.73)$$

where $\hat{P}_{\mathbf{k}\sigma}^\dagger$ ($\hat{P}_{\mathbf{k}_i\bar{\sigma}}^\dagger$) creates a signal polariton at $\mathbf{k}$ (an idler polariton at $\mathbf{k}_i = \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}$), $\mathcal{N}$ is a normalization constant and T_d the detector window. We choose a very wide time window $T_d = 120$ ps, allowing feasible experiments with standard photodetectors. In order to model the density matrix eq. (10.73), we employ the dynamics controlled truncation scheme (DCTS), starting from the electron-hole Hamiltonian including two-body Coulomb interaction and radiation-matter coupling. In this approach nonlinear parametric processes within a third order optical response are microscopically calculated. The main environment channel is acoustic phonon interaction via deformation potential coupling [19, 35].

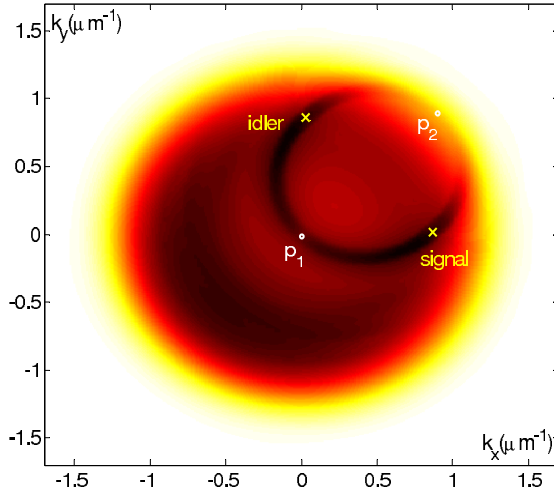


Fig. 10.9 (color online) The simulated spectrally integrated polariton population in $\mathbf{k}$ -space. The parametric process builds up a circle passing through the two pumps (p_1 and p_2), signal and idler polariton states are represented by any two points on the circle connected by a line passing through its center. For illustration the pair of signal-idler polariton modes chosen for entanglement detection are depicted as yellow crosses. The disc-shape contribution centered at the origin is the incoherent population background produced by phonon scattering.

We use a DCTS-Langevin approach [19], with noise sources given by exciton-LA-phonon scattering and radiative decay (treated in the Born-Markov approximation). For mixed-pump processes with arbitrary polarization the Heisenberg-Langevin equations of motion read:

$$\begin{aligned}
\frac{d}{dt} \hat{P}_{\mathbf{k}\sigma} &= -i\tilde{\omega}_{\mathbf{k}} \hat{P}_{\mathbf{k}\sigma} - ig_{|\sigma_1+\sigma_2|} \hat{P}_{\mathbf{k}_1\sigma_1}^\dagger \mathcal{P}_{\mathbf{k}_1\sigma_1} \mathcal{P}_{\mathbf{k}_2\sigma_2} + \hat{\mathcal{F}}_{\hat{P}_{\mathbf{k}\sigma}} \\
\frac{d}{dt} \hat{P}_{\mathbf{k}_i\sigma_i}^\dagger &= i\tilde{\omega}_{\mathbf{k}_i}^* \hat{P}_{\mathbf{k}_i\sigma_i}^\dagger + ig_{|\sigma_1+\sigma_2|} \hat{P}_{\mathbf{k}\sigma} \mathcal{P}_{\mathbf{k}_1\sigma_1}^* \mathcal{P}_{\mathbf{k}_2\sigma_2}^* + \hat{\mathcal{F}}_{\hat{P}_{\mathbf{k}_i\sigma_i}^\dagger} .
\end{aligned} \tag{10.74}$$

In Eq. (10.74) $\mathcal{P}_{\mathbf{k},\sigma}$ are the projections onto the circular basis σ of the coherent pump polariton fields. The explicit expression of the various terms entering the equations of motion (10.74) are:

$$\tilde{\omega}_{\mathbf{k}} = \omega_{\mathbf{k}} - i\Gamma_{\mathbf{k}}^{(\text{tot})}/2 + \sum_{p=1,2} h_{\mathbf{k}}^{(p)} \left| \mathcal{P}_{\mathbf{k}_p}^\pm \right|^2 \tag{10.75}$$

$$\begin{aligned}
h_{\mathbf{k}}^{(p)} = X_{\mathbf{k}} \left[\frac{V}{n_{\text{sat}}} X_{\mathbf{k}_p} (C_{\mathbf{k}_p} X_{\mathbf{k}} + X_{\mathbf{k}_p} C_{\mathbf{k}}) \right. \\
\left. + 2V_{\text{xx}} X_{\mathbf{k}_p} X_{\mathbf{k}_p} X_{\mathbf{k}} \right] .
\end{aligned} \tag{10.76}$$

where X 's and C 's are exciton and photon fraction in the lower polariton branch [19]. The complex polariton dispersion $\tilde{\omega}_{\mathbf{k}}$ includes the effects of relaxation and pump-induced renormalization (shifts $h_{\mathbf{k}}^{(p)}$), g is the nonlinear interaction term driving the mixed parametric processes [19]; summation over the repeated polarization indices σ_1 and σ_2 is assumed and the following selection rule holds: $\sigma_s + \sigma_i = \sigma_1 + \sigma_2$. $h_{\mathbf{k}}^{(p)}$ is the shift induced by the p -th pump due to the parametric process. The damping term $\Gamma_{\mathbf{k}}^{(\text{tot})}$ results from a microscopic calculation including a thermal phonon bath whose details are given in Ref. [19]. V_{xx} is the mean-field while V is the phase-space filling interaction strengths [16, 32, 40, 46, 58]. In general, third order contributions due to Coulomb interaction between excitons account for terms beyond mean-field, including an effective reduction of the mean-field interaction and an excitation induced dephasing. It has been shown that both effects depend on the sum of the frequencies of the scattered polariton pairs [16, 32, 46, 58]. For the frequency range here exploited, the excitation induced dephasing is vanishingly small and can be safely neglected on the lower polariton branch [16, 32, 46, 58]. On the contrary, the matrix elements of the ($J = 0$) counter-circular scattering channel is lower (about 1/3) than that for the co-polarized scattering channel, but certainly not negligible [16, 32, 46, 58]. However, in the pump polarization scheme that we propose, performing the polarization sum in Eq. (10.74), it is easy to see that the counter-circular channel cancels out, as already pointed out. This feature is unique to the present scheme, while all previously adopted pump configurations suffer from the presence of both co- and counter-circular polarized scattering channels. Equation (10.74) is a system of two coupled equations for polariton operators, acting onto the global system and environment state space, thanks to the two additive noise sources $\hat{\mathcal{F}}_{\hat{P}_{\mathbf{k}\sigma}}$, $\hat{\mathcal{F}}_{\hat{P}_{\mathbf{k}_i\sigma_i}^\dagger}$ [19, 45, 74].

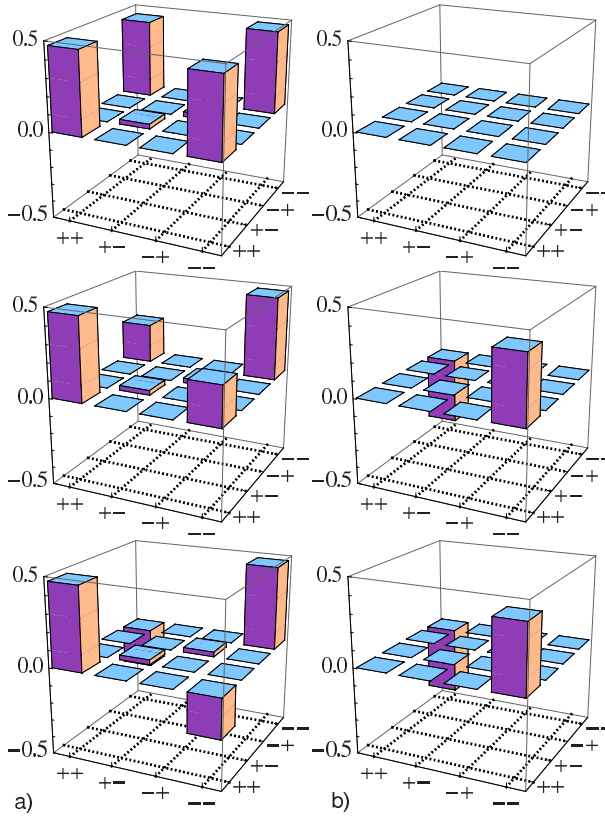


Fig. 10.10 (color online) a(b) Real (Imaginary) part of the density matrix in the tomographic reconstruction according to eq. (10.73) in the linearly cross polarized pump configuration and for different pump polarization in the xy -plane. The first line refers to $\hat{x}$ -polarized ($\theta = 0$), the second line to $\theta = \pi/12$, the third to $\theta = \pi/6$. We point out the different phase relations appearing in the non-diagonal terms directly related to the choice of pump polarization. In the absence of longitudinal-transverse splitting [15] the system is isotropic in the polarization plane, polariton entanglement is independent of the direction of the linear pump polarization angle θ . Indeed the three cases share the same value of entanglement $E(\boldsymbol{\rho}) \simeq 0.7523$.

10.10.2 Results

Figure 10.9 shows a typical pattern of photoluminescence in $\mathbf{k}$ -space we can simulate with our microscopic model. We can neatly distinguish the disc-shape contribution centered at the origin due to the incoherent population produced by phonon scattering from the parametric ring dynamically emerging from the noisy background. The two pumps employed are marked as p_1 and p_2 . Parametrically generated signal and idler polariton states are represented by any two points on the circle connected by a line passing through its center. For illustration the pair of signal-idler

polariton modes chosen for entanglement detection are depicted as yellow crosses. Early experiments in semiconductor microcavities [75, 13] provided promising, though indirect, indications of polariton entanglement. In order to achieve a conclusive evidence of entanglement one has to produce its quantitative analysis and characterization, i.e. a *measure* of entanglement. Among the various measures proposed in the literature we shall use the *entanglement-of-formation* $E(\hat{\rho})$ [70, 71] for which an explicit formula as a function of the density matrix exists [76]. It has a direct operational meaning as the minimum amount of information needed to *form* the entangled state under investigation out of uncorrelated ones [76]. The complete characterization of a quantum state requires the knowledge of its density matrix. Even though its off-diagonal elements are not directly related to physical observables, the density matrix of a quantum system composed by two two-level particles can be reconstructed using the recently developed *quantum state tomography* [69], that has also been exploited in a bulk semiconductor [12]. It requires 16 two-photon coincidence measurements based on various polarization configurations [69]. Exploiting the Wick factorization [44, 77] and the symmetries of the system, the density matrix elements are built up on the signal and idler occupation $N_{s/i\pm} \doteq \langle \hat{P}_{s/i\pm}^\dagger(\tau) \hat{P}_{s/i\pm}(\tau) \rangle$ ($\mathbf{k} = s, i$ mean the chosen signal, idler wave vectors as in Fig. 10.9) and on the two-time correlation functions $\langle \hat{P}_{s+}^\dagger(u) \hat{P}_{i+}^\dagger(v) \rangle$ and $\langle \hat{P}_{s-}^\dagger(u) \hat{P}_{i-}^\dagger(v) \rangle$. Equation (10.74) links $P_{s\bar{\sigma}}^\dagger \leftrightarrow P_{i\bar{\sigma}}$ and $P_{i\bar{\sigma}}^\dagger \leftrightarrow P_{s\bar{\sigma}}$ so that the non-zero elements of the density matrix in our configuration are six:

$$\begin{aligned} \rho_{++,++} &= \rho_{--,--} \\ \rho_{+-,+-} &= \rho_{-+,-+} \\ \rho_{++,-} &= \rho_{--,+}^* \end{aligned} \quad (10.77)$$

We integrate the system of equations eq. (10.74), coupled with the underlying non-equilibrium equations for the noise correlation functions (through time-dependent fluctuation-dissipation relations) [19, 45]. For a generic polarization θ , the populations are independent of θ , $N_{s/i+} = N_{s/i-}$, whereas correlations satisfy $\langle \hat{P}_{s+}^\dagger(u) \hat{P}_{i+}^\dagger(v) \rangle = -e^{i4\theta} \langle \hat{P}_{s-}^\dagger(u) \hat{P}_{i-}^\dagger(v) \rangle$. In Fig. 10.10 a tomographic reconstruction is shown. We point out that different phase relations appearing in the non-diagonal terms of a reconstructed density matrix are directly related to the choice of pump linear polarization θ (see caption of Fig. 10.10). In the absence of longitudinal-transverse splitting [15] polariton entanglement is independent of the angle of the linear pump polarization θ . As Fig. 10.11 shows, there is a non-negligible region of the parameter space where, even in a realistic situation, high entanglement values are obtained. For increasing pump intensities, EOF decays towards zero. This is a known consequence of the relative increase of signal and idler populations [12, 66] – dominating the diagonal elements of $\hat{\rho}$ – with respect to the two-body correlations responsible for the non-diagonal parts, which our microscopic calculation is able to reproduce. We expect entanglement to be unaffected by both intensity and phase fluctuations of the pump laser. The former are negligible according to Fig. 10.12, while the latter only acts on the overall quantum phase of the signal-idler pair state. Different en-

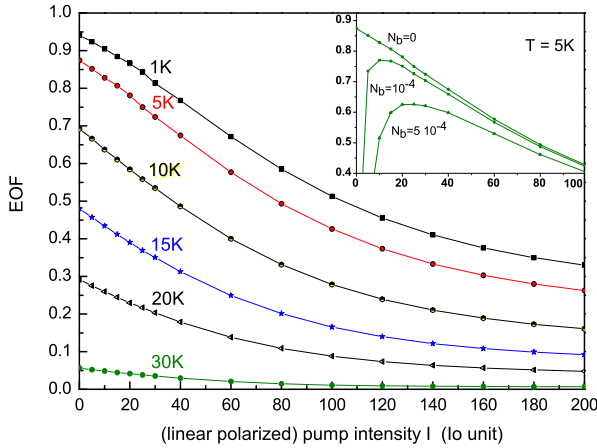


Fig. 10.11 (color online) Dependence of the EOF on pumping intensity. The laser intensity I is measured in units of $I_0 = 21 \text{ photons } \mu\text{m}^{-2}/\text{pulse}$ according to Ref. [43]. In the inset the EOF for the case of $T = 5 \text{ K}$ against pump intensity is depicted for different temperature- and pump-independent noise background N_b representing other possible non-dominant (with respect to acoustic-phonons) noise channels, e.g. the photo-detection system (see text for comments and discussion).

tanglement measures generally result in quantitatively different results for a given mixed state. However, they all provide upper bounds for the distillable entanglement [61], i.e. the rate at which mixed states can be converted into the “gold standard” singlet state. Small EOF means that a heavily resource-demanding distillation process is needed for any practical purpose. Fig. 10.11 shows how a relative small change in the lattice temperature has a sizeable impact on entanglement. As an example, for the pump intensity $I = 15I_0$, increasing the temperature from $T = 1\text{K}$ to $T = 20\text{K}$ means to corrupt the state from $E(\hat{\rho}) \simeq 0.88$ to $E(\hat{\rho}) \simeq 0.24$, whose distillation is nearly four times more demanding. For a fixed pump intensity, Fig. 10.11 shows that, above a finite temperature threshold, the EOF vanishes independently of the pump intensity, i.e. the influence of the environment is so strong that quantum correlation cannot be kept anymore. In physical terms, at about 30K the average phonon energy becomes comparable to the signal-pump and idler-pump energy differences, and the thermal production of signal-idler pairs is activated.

Semiconductors heterostructures are complex systems in which other noise sources are expected. The simplest way to model this additional noise is via the introduction of a constant, temperature- and pump-independent, noise background N_b . This quantity also accounts for the noise background characterizing the photodetection system. In the inset of Fig. 10.11, the dependence of the EOF on N_b is highlighted. In our simulation, the quantity N_b causes EOF to vanish in the limit of low pump intensity. A value of N_b of about 10^{-4} is realistic, as suggested by experiments [13] showing that it is considerably smaller than the PL-noise studied here. From inspection of the density matrix (in the $(++, +-, -+, --)$ circular-

polarization basis), for pump intensity $I \rightarrow 0$, if the correlation dominates, we have as limiting case a triplet pure state

$$\rho \rightarrow \frac{1}{2} \begin{pmatrix} 1 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 1 \end{pmatrix}. \quad (10.78)$$

On the other hand, if the population dominates, the state becomes separable $\rho \rightarrow \frac{1}{4}\mathbb{I}$, ($\mathbb{I}$ the unit matrix). At leading order in I , the two contributions are comparable, and $1 - E(\hat{\rho})$ is determined solely by the ratio between incoherent and parametric scattering rates. However, for ultrafast pulsed excitations, necessary for QIC applications, things are more complex and this simplified analysis fails. Indeed, the dynamical interplay between noise and parametric processes is the principal ingredient of interest and physical explanations with quantitative predicting character need to take it properly into account. For nonzero N_b , instead, the leading term in the population is constant. This explains the behaviour of EOF as $I \rightarrow 0$. Mathematically speaking, for any finite value of N_b , we have a separable state in the limit $I \rightarrow 0$. As seen in the inset of Fig. 10.11, a finite N_b affects only the range of very low pump intensity I . Figure 10.12 displays the time-resolved signal/idler occupations calculated at $T = 5K$ for three different excitation densities. The dashed lines describe the PL contributions to the occupation. The figure clearly shows that at low excitation density the detected intensity in the signal/idler channels arises mainly from PL. Nevertheless the obtained EOF for these intensities is very high, contrarily to intuition, but in agreement with recent results [66]. This result puts forward the robustness of pair correlations and entanglement that can be evidenced, even when noise is the dominant contribution to one-body properties.

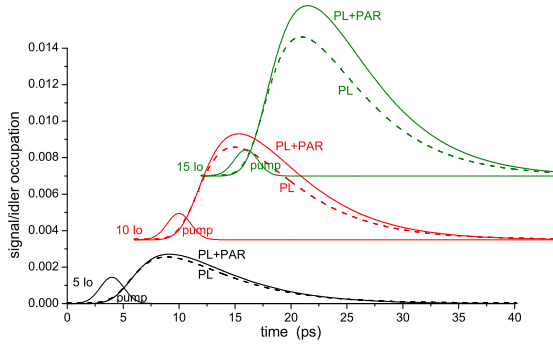


Fig. 10.12 (color online) Signal/idler occupations (dashed: PL only; full: PL+parametric) vs time for three excitation densities ($I = 5 I_0$, $I = 10 I_0$ and $I = 15 I_0$) for $T = 5K$ are shown. The shape of the pump pulse is depicted for reference.

10.11 Outlook

In this chapter we reviewed quantum optical effects with excitonic polaritons in both bulk semiconductors and in cavity embedded QWs. Although, the excitonic polariton concept, introduced in 1958 by J.J. Hopfield [78], was based on a full quantum theory including light quantization, the investigations of the optical properties of excitons developed mainly independently of quantum optics. The first manifestation of excitonic quantum-optical coherent dynamics was observed experimentally 20 years later [38] exploiting the resonant hyper-parametric scattering. The possibility of generating entangled photon pairs by this process was theoretically pointed out only lately [24]. This theoretical prediction was based on a microscopic quantum theory of the nonlinear optical response of interacting electron systems relying on the dynamics controlled truncation scheme [10] extended to include light quantization [16, 24]. The experimental evidence for the generation of ultraviolet polarization-entangled photon pairs by means of biexciton resonant parametric emission in a single crystal of semiconductor CuCl has been reported [12] in 2004. The demonstrations of parametric amplification and parametric emission in SMCs [49, 57], together with the possibility of ultrafast optical manipulation and ease of integration of these micro-devices, have increased the interest on the possible realization of nonclassical cavity-polariton states. In 2005 an experiment that probes polariton quantum correlations by exploiting quantum complementarity has been proposed and realized [13].

These results have unequivocally proven that despite these solid state systems are far from being isolated systems, quantum optical effects at single photon level arising from the interaction of light with electronic excitations of semiconductors and semiconductor nanostructures were possible. The control over the interaction between single photons and individual optical emitters is an outstanding problem in quantum science and engineering. In the last few years substantial advances have been made towards these goals by achieving the strong coupling regime for a single quantum dot embedded in a high-Q microcavity [79, 80]. The quantum nature of this strong coupled system has been demonstrated [81]. Of great interest is also the interaction of quantum emitters with surface plasmon modes supported by metallic nano-particles [82, 83]. These hybrid metal-semiconductor systems can display quantum optical effects promising for realization of sub-wavelength devices.

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Chapter 11

Spontaneous coherence within a gas of exciton-polaritons in Telluride microcavities

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Abstract Microcavity exciton-polaritons are the eigenstates resulting from strong light-matter coupling in high quality monolithic semiconductor microcavities. Owing to their mixed photonic and excitonic nature, polaritons are Bose particles of very light mass and short lifetime that can interact with their environment, forming a new class of Bose gas. In spite of their short lifetime, a polariton gas can show Bose-Einstein condensation or polariton lasing depending on the experimental conditions. The properties of these coherent states of polaritons are unique in many respects. In this chapter we rely on a comprehensive set of experimental and theoretical works carried out this last decade to give a detailed description of polariton coherent states. The influences of finite lifetime, disorder and interaction with the environment are addressed, and the analogies and differences between polariton condensate, polariton lasing, equilibrium Bose gas and photon lasing are outlined and discussed.

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11.1 Introduction

Exciton-polaritons are the elementary excitations of a semiconductor microcavity operating in the strong coupling regime. They have a mixed exciton-photon nature which results from the strong interaction between quantum well excitons and cavity confined light. In the dilute regime, since both exciton and photon feature integer spin, polaritons are bosonic particles. In the vast zoology of Bose gases, they present unique characteristics: due to their photon component, they are extremely light particles (4 orders of magnitude lighter than free electrons) and their lifetime is finite due to radiative recombination at the microcavity mirror/air interface. Their light weight is a striking advantage over more massive bosons: since the thermal de Broglie wavelength scales like $1/\sqrt{m}$, quantum degenerate regime where single wavefunctions start to overlap can show up in a polariton gas at temperatures as high as room temperature.

On the other hand their very short lifetime changes dramatically the picture with respect to equilibrium Bose gases. The polariton gas needs to be constantly replenished by an external "pump" to compensate for the losses and establish a steady state. State of the art microcavities exhibit a polariton life time of up to 10 ps. This lifetime is too short to achieve thermal equilibrium with the lattice by scatterings with optical and acoustic phonons. However, at density high enough to form a polariton condensate, polariton-polariton and polariton-exciton scattering rates can be large enough compared to the life time to establish thermal equilibrium within the polariton gas, with a well defined temperature different from that of the lattice. Thus any realistic theoretical description of the polariton system should take this non-equilibrium character into account. In addition to be responsible for this driven-dissipative situation, the polariton radiative recombination results in a very useful property: the emitted photon field conserves most observables describing the polariton field : density, energy, momentum, polarization, correlations (first and second order have been studied so far). Thus the polariton gas inside the microcavity can be entirely characterized by performing optical measurements. This direct access to physical quantities is a strong advantage over other quantum degenerate systems like superfluids, superconductors, or cold atom gases.

The first theoretical discussion on the strong potential of exciton polaritons for the realization of unusual quantum degenerate Bose gases was proposed by A. Imamoglu and co-workers in 1996 [18]. A first clear observation of bosonic stimulation within a polariton gas was obtained by L.S. Dang and co-workers in 1998 [12] in a Telluride-based microcavity, where the exciton is robust enough to sustain the carrier density at the stimulation threshold. Indeed, with a lower excitonic binding energy like in Arsenide-based microcavities it is more difficult to experimentally discriminate between regular photon lasing in the weak coupling regime and polariton condensation in the strong coupling regime [35, 8, 44, 13]. Finally, unambiguous demonstrations of polariton Bose-Einstein condensation (BEC) were reported in 2006 [20] and afterwards [6, 54]. In this chapter based on recent experimental findings and theoretical development, we will address in details some crucial

issues: formation of the polariton gas, momentum distribution, polariton BEC and polariton lasing vs. photon lasing, spatial properties, and fluctuations.

11.2 Formation and steady state of a polariton gas

The excitation of polaritons in a microcavity is realized optically. To achieve spontaneous formation of polariton coherence, caution must be taken not to introduce any coherence in the system via the excitation laser. Thus non-resonant excitation (typically 100meV above the polariton states) is in general carried out. Importantly, this excitation scheme also allows for thermodynamics to play an important role in the mechanism of polariton condensation.

Objects similar to a polariton condensate can be realized with different excitation techniques. For instance, in the so-called parametric oscillation scheme, a well defined polariton state is optically driven with a laser beam [43, 46]. Above an intensity threshold, pumped polaritons undergo stimulated inelastic scatterings into the signal (usually the ground state) and idler polariton states. In this process a well defined phase emerges spontaneously for the signal and idler fields independently from the laser phase [58]. In this sense the behavior of the signal population through threshold resembles a Bose gas undergoing condensation; on the other hand, thermodynamics plays no role whatsoever in its formation. In the non-resonant scheme (summarized in Fig.11.1.a), the laser beam excites hot free electron-hole pairs. The free particles relax the main part of their excess energy by several very fast emissions of LO phonons in the lattice. A hot exciton gas is thus formed [52]. For this population to relax into the polariton states of lowest energy, a few tens of meV below, acoustic phonon emission is the only possible channel. This relaxation mechanism is very inefficient due to the very steep energy dispersion of polariton states. This problem, known as the "relaxation bottleneck" was well identified both experimentally [49, 34] and theoretically [50], using a Boltzmann description of the dynamics. The left panel of Fig.11.1.c shows a measurement of the polariton emission along the low energy part of the lower polariton branch under weak excitation (low polariton density) and negative exciton-cavity detuning. A pronounced population bottleneck is observed: the high energy polariton states are much more populated than the lower energy ones.

At high excitation intensity, i.e. at high exciton density, the relaxation bottleneck can be overcome by exciton-exciton inelastic scatterings. It was predicted in 2002 by Porras and co-workers [37] that this relaxation channel could build up a polariton ground state occupancy high enough to trigger bosonic stimulation of the relaxation (cf. Fig11.1.b). This prediction is in good agreement with the experiments : upon increasing excitation intensity, a first nonlinearity is in general observed, where the polariton ground state density increases quadratically with respect to the injected exciton density. This behavior is shown in Fig.11.1.d : below threshold, the ground state polariton population vs excitation laser intensity is much better fit with a second order polynomial than with a line. This quadratic contribution is a sig-

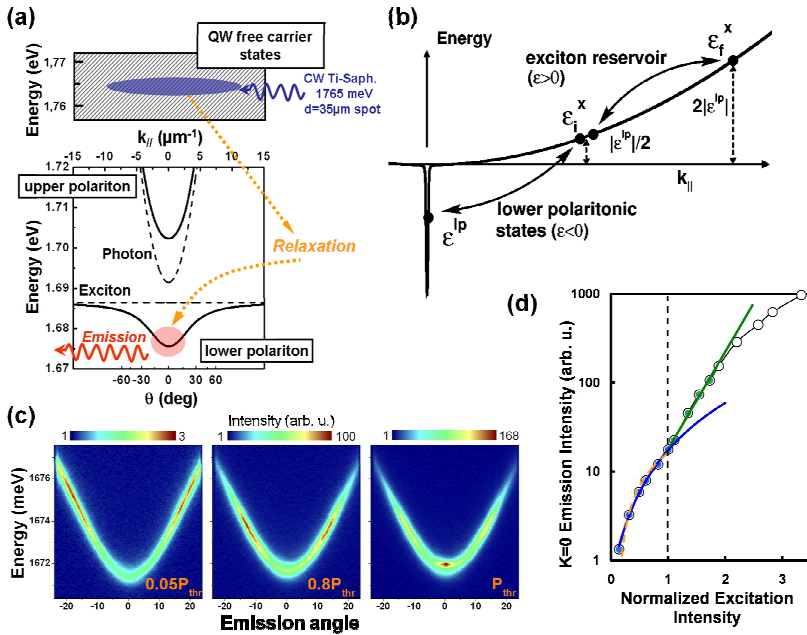


Fig. 11.1 (a) Schematic representation in a (Energy, momentum) plane of the polariton gas excitation by a non-resonant laser beam. The chosen energy scale is typical of polariton energies in Telluride-based microcavities. (b) Picture reproduced from [37] : schematic representation in a (Energy, Momentum) plane of the exciton-exciton scattering process used in the model of Porras and co-workers as a possible channel for polariton condensation. The Momentum scale is much larger than in (a). (c) Measurement in a (Energy, Momentum) plane of the polariton emission in a Telluride-based microcavity featuring 26 meV Rabi splitting (16 CdTe quantum wells) and -1 meV exciton-photon detuning. The left panel is obtained far below the degeneracy threshold (P_{thr} is the excitation laser power at the degeneracy threshold), the middle one is close to the threshold, and the right one is obtained at threshold. (d) is a measurement of the ground state polariton population upon crossing the degeneracy threshold (vertical dashed line). Below threshold, a linear (dashed orange line) and second order polynomial (blue solid line) fits of the data (blue circles) are shown and highlight the contribution of a quadratic nonlinearity. Above threshold (green circles) and before the saturation regime (open circles), an exponential function fits well the data (green solid line).

nature of exciton-exciton scattering events [37]. This nonlinear relaxation channel doesn't affect only the ground state population : in the middle panel of Fig.11.1.c the pump intensity has been increased with respect to the left one to reach the regime where exciton-exciton scattering contribution is significant. The relaxation bottleneck is found to be partially suppressed and the overall population is shifted toward lower energy. Upon increasing further the pump intensity, the stimulation threshold is reached. As shows the measurement of Fig.11.1.d, a much stronger nonlinearity starts (exponential dependence with the pump intensity instead of quadratic) due to bosonic stimulation of the polariton relaxation toward the ground state [12]. In this regime, the polariton condensate is formed and dominates the whole popula-

tion. This situation corresponds to the measurement shown in the right panel of Fig.11.1.c, where the red spot at the bottom of the lower polariton branch is the polariton condensate.

11.3 Momentum distribution, polariton thermalization

In the theory of Bose gases, two distinct classical regimes can be identified. In the first one, the bosons behave as classical particles, undergoing scatterings. This regime can be described with a Boltzmann equation for the occupation numbers $n_{\mathbf{k}}$ of the momentum states $\mathbf{k}$. The second classical regime is achieved when most of the particles occupy a single quantum state. Then, this macroscopic wavefunction is a coherent field which resembles very much a classical wave like a laser field, and its behavior is governed by a wave equation : the time-dependent Gross-Pitaevskii equation. A unified theory that simultaneously describes all the physical phenomena in these two regimes is at present not available. Below, we will show that the Boltzmann description can successfully describe the experimentally observed momentum distribution of polaritons, and in Section 11.5, we will explain the observed spatial phenomena in the condensate state within the Gross-Pitaevskii framework.

The most complete quantitative comparison between experiments and theory based on the Boltzmann equation was recently published in Ref. [21], where the issue of thermalization was investigated. The theoretical model includes relaxation mediated by acoustic phonons and exciton-exciton scatterings. A hot exciton reservoir (at a temperature $k_B T \simeq E_{LO}$ where $E_{LO} = 21.3 \text{ meV}$ in CdTe) is assumed due to very fast LO-phonon relaxation of the photocreated free carriers.

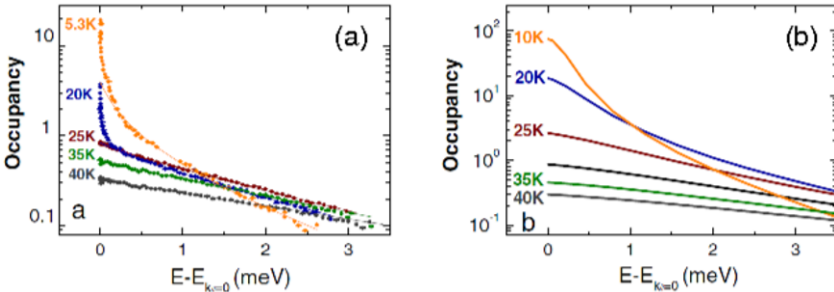


Fig. 11.2 Taken from [21]. (a) Measured polariton occupancy versus energy for decreasing temperature (40K to 5.3K) and for polariton density kept constant. The BEC phase boundary is crossed between 25K and 20K. The solid lines are fits using a Bose-Einstein distribution. (b) Calculation of the polariton occupancy using a Boltzmann approach.

At positive and close to zero photon-exciton detunings and above the critical density, the polariton condensate builds-up in the ground state ($k = 0$) and the non-condensed polaritons follow a Maxwell-Boltzmann distribution, characteristic of

thermal equilibrium. It should be noted that in systems out of equilibrium, dynamical effects can sometime result in a similar distribution shape. For instance it has been observed in a microcavity operating in the VCSEL (conventional photon lasing) regime [4]. However, as opposed to VCSELS, and in agreement with equilibrium Bose gas behaviour, the polariton BEC can be fully driven by temperature alone. This is shown in Fig.11.2: In this experiment, the polariton density is kept constant and the lattice temperature is decreased : from 40K to 25K a Maxwell-Boltzmann distribution is observed with decreasing characteristic temperature. Below 25K a Bose-Einstein-like low energy-peaked distribution develops, characteristic of the transition into the condensate regime. This behavior is well captured by the model, and proves directly a point of major importance : in the proper experimental condition polariton BEC can be driven by thermodynamics. A very different situation is observed at negative photon-exciton detuning. Close to the threshold, the non-condensed polaritons do not follow a Boltzmann distribution anymore, i.e. the bottleneck effect is still effective even above threshold. This situation is for instance that of Fig.11.1.c. It is also observed that the threshold density decreases with increasing temperature [21], which is not consistent with the behavior of a Bose gas at thermal equilibrium. In this regime, in analogy with the non-equilibrium character of lasers, we rather speak of "polariton lasing" instead of polariton BEC. Factors that play an important role to explain this phenomenology are the steeper polariton dispersion (lower density of states), the larger energy difference between the exciton reservoir and the polariton ground state (more energy should be dissipated to cool down) and the shorter polariton life time due to the larger photon fraction of the ground state. Finally there exists an intermediate situation, where the polariton distribution is non-thermal close to the threshold but develops a Bose-Einstein-like distribution deeper in the condensed regime.

To conclude this section it is important to keep in mind that even when thermodynamic equilibrium is reached in the above defined sense (i.e. when the thermalization time is shorter than the lifetime), it is never achieved *stricto-sensu*: the system is always in a driven-dissipative situation with comparable timescale of losses and pumping dynamics.

11.4 Similarities and differences between polariton condensation, polariton lasing and conventional photon lasing

Textbook Bose Einstein condensation is a phase transition that is entirely driven by quantum statistics, where interactions play only a secondary role. It occurs when a gas of bosons with mass m and density n is cooled below the critical temperature $T_c \propto n^{2/3}/m$ or when at fixed temperature the density is increased above the threshold density $n_c \propto (mT)^{3/2}$. The transition is marked by the onset of a macroscopic occupation of a single particle state and as a consequence long range spatial coherence. Even though BEC was predicted by Einstein to occur in a non-interacting

system, any practical realization relies on interactions, owing to the trivial observation that the momentum distribution of a gas without any interactions does not evolve and no macroscopic occupation of a single particle state may spontaneously form. Interactions let the gas evolve to its thermal equilibrium state, where the phase transition occurs.

Because the same $U(1)$ symmetry is broken when the BEC or lasing thresholds are crossed, there should be a smooth crossover between both cases and no clear cut distinction between them is possible. Starting with a BEC of stable bosons, we can go toward the lasing regime by introducing single particle losses. They can be compensated by some gain that adds new incoherent particles to the gas. When the boson life time becomes shorter than the thermalization time, thermal equilibrium is lost and the physics of a laser is recovered. Polariton BEC in semiconductor microcavities occurs in this crossover regime where, depending on the experimental conditions, laser or BEC physics is relevant. A formal description of this smooth crossover is presented chapter 12.

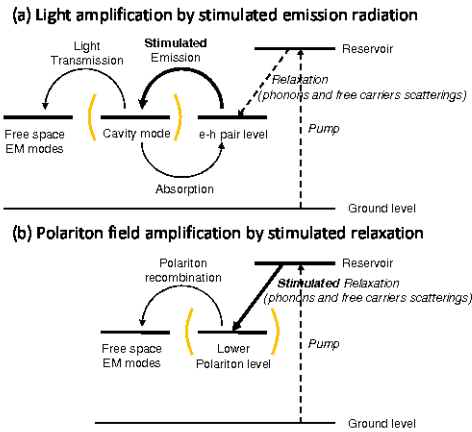


Fig. 11.3 Schematic representation of the principle of a three level conventional photon laser (a) and of a three level polariton laser

We want to stress that despite the formal analogies, there are very important differences between conventional lasing operation in semiconductor microcavities (VCSEL), polariton condensation and polariton lasing. In the previous section, we have seen that this out-of-equilibrium Bose gas could be in internal equilibrium or not, depending on the experimental conditions. In the first case, the phase transition can be driven both by increasing the polariton density or by decreasing the temperature. This last possibility is forbidden for a laser in which thermodynamics cannot play that role. In the second case, i.e. in the polariton lasing regime, the similarity with a laser is closer since for both, the thermodynamic role is negligible in the phase transition.

Still, two fundamental differences are always present :

- For photon lasers, the light-matter interaction is in the weak coupling regime, and the bosonic field which is amplified by Bose stimulation is the bare electromag-

netic field. For polariton lasing, we are in the strong coupling regime, dealing with a mixed exciton-photon (polariton) field.

- The mechanisms for photon and polariton stimulation are very different. They are summarized in Fig.11.3. In the first case, lasing takes place when the balance between absorption and emission in the gain medium (free electron-hole plasma in the case of VCSELs) is achieved. For a microcavity in the strong coupling regime, population inversion is not required because the system is always transparent for polaritons even with the pump off. Thus the only condition for polariton stimulation to occur is to achieve the balance between polariton creation and polariton losses through the cavity mirrors.

11.5 Spatial properties

11.5.1 Gross-Pitaevskii equation

The above discussion on polariton dynamics illustrates the success of a Boltzmann approach to account for the dynamics of the high energy excitations of the polariton gas (energies above the condensate's) including bosonic stimulation and scattering mechanisms. However, it is a completely incoherent approach which fails to describe the coherent phenomena that take place when a significant fraction of the population is condensed into the ground state. Especially the effect of spatial inhomogeneities are not well described, because in the physics of spatially inhomogeneous polariton condensates, the superfluid velocity $v_c = \nabla\theta/(\hbar m)$ (where θ is the phase of the order parameter) plays an important role. On the other hand, when the majority of particles occupy a single particle state as for Bose-Einstein condensate, the behavior is well captured by a fully coherent approach in which the system is approximated by a single "classical" wavefunction ψ of well defined phase and amplitude, and its evolution is governed by the time-dependent Gross-Pitaevskii (GP) equation

$$i\hbar \frac{\partial}{\partial t} \psi(\mathbf{r}) = \left[-\frac{\hbar^2}{2m} \nabla^2 + V_{ext}(\mathbf{r}) + g|\psi(\mathbf{r})|^2 \right] \psi(\mathbf{r}) \quad (11.1)$$

where g is the mean field interaction, m the particle mass and $V_{ext}(\mathbf{r})$ the external potential. This equation neglects all quantum fluctuations of the bosonic field, but correctly describes the nature of the long wavelength excitations and the spatial structure of the condensate in external potentials. On the other hand, for higher energy excitations like the thermal component surrounding the condensate, the coherence is negligible. Hence, their dynamics are better captured by a Boltzmann equation.

The GP equation 11.1 describes a lossless field. Thus for the proper description of polariton condensate it needs to be generalized to account for condensate radiative losses and replenishment by the exciton reservoir. The simplest phenomenological description of this mechanism is to consider the exciton reservoir as a gain medium

for the polariton condensate. The polaritons then evolve according to a generalized GP equation :

$$i\hbar \frac{\partial}{\partial t} \psi(\mathbf{r}) = \left\{ -\frac{\hbar^2}{2m} \nabla^2 + \frac{i\hbar}{2} [R[n_R(\mathbf{r})] - \gamma] + V_{ext}(\mathbf{r}) + \hbar g |\psi(\mathbf{r})|^2 + V_R(\mathbf{r}) \right\} \psi(\mathbf{r}), \quad (11.2)$$

where γ is the polariton radiative rate. Note that this equation can also be derived as the limit of a more general description of the condensate as explained in chapter 12 section 4.4. At the simplest level, the corresponding gain rate $R[n_R]$ can be described by a monotonically growing function of the local density $n_R(\mathbf{r})$ of reservoir excitons [37]. At the same time, the reservoir produces a mean-field repulsive potential $V_R(\mathbf{r})$ that can be approximated by the linear expression $V_R(\mathbf{r}) \simeq \hbar g_R n_R(\mathbf{r}) + \hbar \mathcal{G} P(\mathbf{r})$, where $P(\mathbf{r})$ is the (spatially dependent) pumping rate and $g_R, \mathcal{G} > 0$ are phenomenological coefficients to be extracted from the experiment. The time-evolution of the excitonic reservoir coupled to the polariton macroscopically occupied wave function is

$$\dot{n}_R(\mathbf{r}) = P(\mathbf{r}) - \gamma_R n_R(\mathbf{r}) - R[n_R(\mathbf{r})] |\psi(\mathbf{r})|^2. \quad (11.3)$$

where γ_R is the reservoir loss rate. Note that this set of coupled equations (11.2) and (11.3) has exactly the same structure as the equations commonly used to study spatio-temporal dynamics in VCSELs [25]. In the limit where γ_R is much faster than any other frequency in the problem, this set of equations can be reduced to a complex Ginzburg-Landau equation (CGLE), widely studied in the context of planar laser physics and in pattern formation out of equilibrium in general [11]:

$$i\hbar \frac{\partial}{\partial t} \psi(\mathbf{r}) = \left\{ -\frac{\hbar^2}{2m} \nabla^2 + i(\varepsilon - r |\psi(\mathbf{r})|^2) + V_{ext}(\mathbf{r}) + \hbar g |\psi(\mathbf{r})|^2 \right\} \psi(\mathbf{r}), \quad (11.4)$$

ε is the effective gain (gain minus losses) and r is the gain saturation parameter. The steady state density n_s is determined by the balance between the pumping and dissipation: $n_s = \varepsilon/r$. This equation was applied to the study of polariton condensates in external potentials by Keeling and Berloff [22]. It is in fact instructive to rewrite the extended GP equation with the order parameter in polar coordinates $\psi = \sqrt{\rho} e^{i\theta}$:

$$\frac{\partial}{\partial t} \rho = [R[n_R] - \gamma_c] \rho - \nabla \cdot (\rho \mathbf{v}_c) \quad (11.5)$$

$$\hbar \frac{\partial}{\partial t} \theta = \frac{m}{2} v_c^2 + \frac{\hbar^2}{2m} \frac{\nabla_{\mathbf{r}}^2 \sqrt{\rho}}{\sqrt{\rho}} + V_{ext} + \hbar g \rho + \hbar g_R n_R + \hbar \mathcal{G} P \quad (11.6)$$

$$P = \gamma_R n_R + R[n_R] \rho, \quad (11.7)$$

Equation (11.5) is the continuity equation, that takes into account the gain and losses, as well as the convection due to the coherent flow of velocity $\mathbf{v}_c = \nabla \theta / (\hbar m)$. Equation (11.6) describes the time evolution of the phase. In the steady state, the left hand side reduces to the condensate frequency ω_c and the equation then expresses a kind of spatial energy conservation. The condensate frequency is a sum of kinetic

(the first two terms) and potential energy. Then if the potential energy is lower at some points of space, this should be compensated by the kinetic energy. This is important for understanding the shape of condensates in inhomogeneous geometries.

11.5.2 Small size condensate

In the early attempts to achieve degenerate states of polaritons, a puzzling observation was reported (cf. Fig.11.4.b) when a dense polariton gas was excited on a very small area (~ 1 micron diameter); above a critical density, a degenerate polariton gas, driven by a strong bosonic stimulation, builds up into the shape of a thin ring (of well defined momentum magnitude) in momentum space [39]. The emergence of correlation in momentum space within this ring at the critical density was clearly identified by interferometric measurements. The mechanism underlying this phenomenon remained unclear until recently, when a mean-field approach successfully retrieved this behavior [59]: this peculiar condensate ring shape in momentum space is in fact a consequence of the ballistic expansion of the polariton condensate out of the excitation region. Indeed the inhomogeneous polariton density produces an anti-trapping potential, due to polariton-polariton and polariton-carrier repulsive interactions. The polariton condensate created in the center of the excitation spot experiences this strong potential gradient which pushes polaritons out of the dense area, thus acquiring a well defined kinetic energy, identical in every directions. The underlying conservation of the energy, which governs this mechanism, is formally expressed by Eq. (11.6). This behavior is well reproduced by the calculation (cf. Fig.11.4.d) using the parameters used in the experiment. When the size of the excitation laser is increased, the potential gradient is reduced and the polaritons do not have enough time to acquire a significant kinetic energy and leave the dense area within their lifetime. Then the condensate takes a more usual shape, peaked around the zero momentum state. Nonetheless, due to the non zero polariton acceleration, the condensate still presents a significant momentum broadening exceeding the one due only to its finite size. The corresponding experimental and theoretical results are shown in Fig.11.4. To really suppress this pump induced outward polariton flow, an excitation spot with a top-hat intensity profile and with a large diameter has to be used, as in the measurement in Fig.11.1.c.

The dependence of the momentum distribution on the spot size is a good illustration of the consequences of the non-equilibrium character of the polariton condensate. At thermal equilibrium, the time-reversal of the Gross-Pitaevskii equation guarantees that always a real solution for the order parameter can be found. Then, the momentum distribution is always centered around $k = 0$.

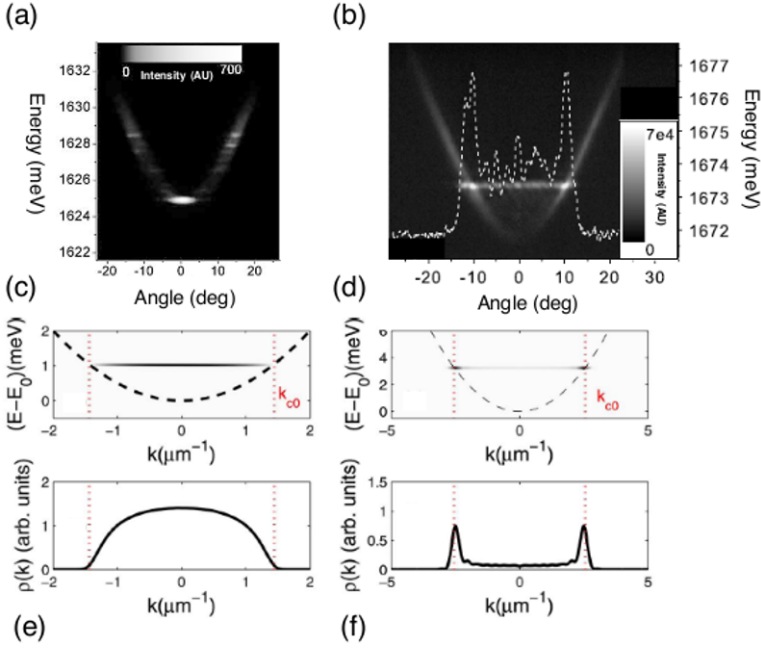


Fig. 11.4 Polariton emission measured in the (momentum, Energy) dispersion plane in the condensate regime for (a) a large excitation spot of $\sim 20\mu\text{m}$ diameter, (b) a small excitation spot of $\sim 1\mu\text{m}$ diameter (taken from [39],[38]). In (a) the condensate is the bright white spot at the lowest energy. In (b) the condensate is the flat feature at 1673.3meV. The dashed line is a plot of its density versus momentum. The condensate density is calculated for the experimental conditions (a) and (b) using the generalized Gross-Pitaevskii equation. The results are shown in (c) and (d) respectively, in the dispersion plane (the condensate density is represented by shades of gray) and in (e) and (f) respectively where the condensate density versus momentum is plotted (taken from [59]).

11.5.3 Polariton condensate in disordered environment

Epitaxial semiconductor microcavities in the strong coupling regime exhibit disorder of both excitonic and photonic kinds. The first one originates from alloy composition inhomogeneity and quantum well thickness fluctuations. The resulting disordered potential experienced by the exciton has the following typical figures : a few meV amplitude and 10-100nm correlation length [42]. Cavity photons are also subject to a disordered potential which results from nanometer-scale thickness fluctuations of the whole cavity. Typical figures describing this disorder are a fraction to a few meV amplitude and correlation length in the several microns range, i.e. much longer than the excitonic one. Reducing further this disorder is a true technological challenge since for a 2λ cavity thickness, they correspond approximately to fluctuations of a single atomic monolayer.

Early discussions on the origin of polariton inhomogeneous broadening assumed that excitonic disorder was the main cause. However, it was later established both

experimentally and theoretically [55, 40] that the very light mass of the polaritons compared to the excitonic one averages out this disorder and causes a motional narrowing of the polariton spectral line. Thus, the excitonic disorder can be in general disregarded in the strong coupling regime as long as the Rabi splitting is large compared to the disorder amplitude. On the other hand, the photonic disorder features a much longer correlation length, comparable to the polariton coherence length. In this case, the disorder is not averaged out by motional narrowing, and leads to significant localization and inhomogeneous broadening of the polaritons that must be taken into account [41]. Typical values of disordered potentials experienced by polaritons in Telluride-based microcavities, measured by spectrally resolved micro-photoluminescence, are 1-4 microns for the correlation length and 0.8 meV for the mean amplitude. In Arsenide-based microcavity, the observed cavity-induced disorder is in general shallower and more structured : spatially regular patterns have been reported several times in the far field of Rayleigh scattering, corresponding to a regular grid in direct space with periods of tens microns due to the lattice mismatch at the interfaces of the AlGaAs layers present in the Bragg mirrors. [16, 29].

11.5.3.1 Mode locking, role of the nonlinearities

A defining property of Bose-Einstein condensation is the build-up of spatial long range correlations above the condensation critical density. On the other hand disorder is a notorious enemy of long range coherence. Still, in a polariton condensate, the correlations are often found to extend much further ($\sim 20\mu m$) than the disorder correlation length. At thermodynamic equilibrium, it is well known that disordered potentials can have dramatic effects on Bose-Einstein condensation and can drive the Bose gas into the Bose glass phase, without long range order (exponential decay of the spatial coherence [15]). Actually the disorder potential will induce a spatial modulation of the condensate density. Thus adjacent high-density regions may be weakly coupled because of the high potential barriers between them. Quantum fluctuations can then be sufficient to destroy the long range coherence. For polariton condensates, the Bose glass phase was theoretically predicted by Malpuech *et al.* [31] by using an equilibrium Gross-Pitaevskii equation and physical parameters of CdTe microcavities. However, upon including the effect of driving and dissipation, a different picture is obtained. It turns out that the condensate does not necessarily oscillate at a single frequency, but the steady state may involve the macroscopic occupation of several states at different frequencies. In disordered structures, localized eigenstates are found at different places with different energies. Above the condensation threshold, several of them become macroscopically occupied. Usually the condensate states are different from the single particle eigenstates, because the real and imaginary nonlinearities in Eq. 11.2 or Eq. 11.4 modify the linear wave functions. Under certain conditions, the nonlinearities give rise to a spatially extended linear combination of single particle eigenstates that oscillates at a single frequency.

A detailed experimental study of the disorder influence on the polariton condensation has been carried out in [3]. The attention was focused on the emission spectrum from neighboring maxima in the condensate density. It was found that depending on the actual realization of the in-plane disorder (in particular the peak to valley energy difference), one single condensate exhibiting long range correlations and a single frequency could be formed (shallow disorder case : Fig.11.5.b), or several independent small size condensates each with a close but different frequency are formed (larger amplitude disorder case : Fig.11.5.d). Interestingly, although an approach based on a generalized GP equation like the one of Eqns.11.2 and 11.3 successfully accounts for this behavior of polariton condensates [57], this phenomenon of frequency synchronization is a characteristic feature that can be found in totally different ensembles of coupled nonlinear oscillators. Synchronized blinking of fireflies, synchronization of cardiac pacemaker cells, laser mode locking are all contained in this framework [56] and their macroscopic behavior is governed by the Adler equation [1]

$$\frac{d\theta}{dt} = \delta + g \cdot \sin \theta \quad (11.8)$$

where θ is the relative phase between the oscillators, δ their relative frequency detuning, and g the nonlinearity magnitude. In our physical situation, the oscillators are polaritons at different localization sites, the nonlinearity is the polariton-polariton interaction, and the coupling between different sites is mediated by polariton coherent tunneling, i.e. Josephson currents of polaritons [57].

Subsequent experiments revealed an even richer phenomenology. Using a laser diode instead of a Ti:sapphire laser for the excitation laser, the excitation intensity noise was significantly reduced. Then it appeared that the "single large size condensate" situation of the previous experiment actually corresponds to the large size multimode condensate as described above. Moreover each mode linewidth was found to be as low as a few tens of μeV [23]. The dramatic dependence of the condensate linewidth on the excitation laser (Ti:Sa or semiconductor diode) is explained by the different time scales of their intensity fluctuations. In the semiconductor diode, intensity fluctuations are on the scale of a few ps, so that they are washed out by the slow exciton dynamics. The intensity fluctuations in the larger Ti:Sa laser are much slower (tens of ns) so that the density (and associated blue shift) vary adiabatically with the laser intensity. As a consequence the time integrated spectra are smeared out into a single inhomogeneously broadened line. The emission spectrum observed in these experiments was predicted by studies of the CGLE [14] with external potentials, and can be qualitatively well understood by numerical simulations based on the extended Gross-Pitaevskii equation [57]. In Figs. 11.6.a and 11.6.b, we show a measurement in the (momentum, Energy) plane of the polariton photoluminescence resolved in two orthogonal linear polarizations. The corresponding integrated spectrum is shown in Fig. 11.6.c. It consists of six peaks, each of them corresponding to an independent macroscopically occupied mode. The corresponding real space distributions are shown in Fig. 11.7. Theoretical analysis has shown that the lowest energy condensate state is very similar to the lowest single particle eigenstate within the excitation spot. The other modes are formed out of linear combinations of single

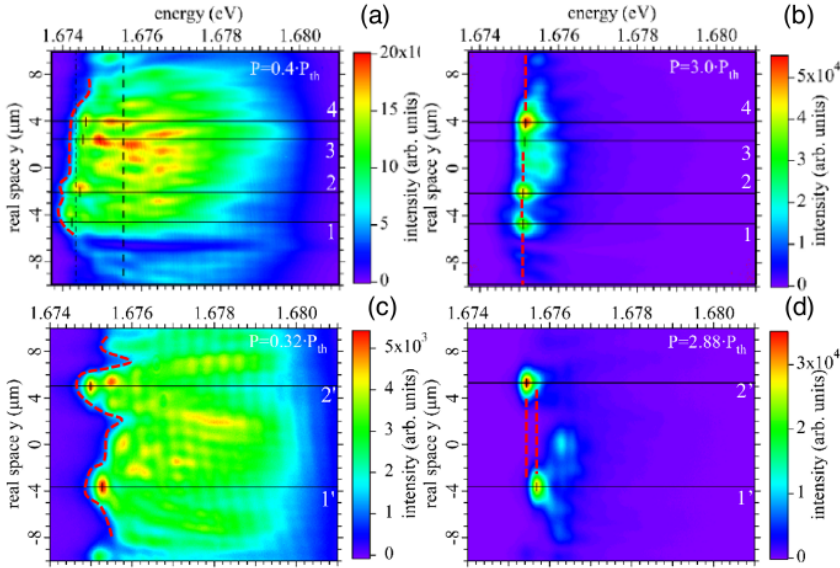


Fig. 11.5 Spectrally-resolved micro-photoluminescence of a polariton gas. (a) and (c) in the low density regime : The low energy emission cut-off provides a good approximation of the local polariton ground state energy. The dashed red line materializes the spatial fluctuations of the latter : the disorder has a larger amplitude in (c) than in (a). (b) and (d) are obtained above the critical density. (b) shows frequency synchronization : points 1,2,3,4 have reduced their respective detunings to 0. In (d) a single frequency could not be found : detunings at points 1' and 2' are reduced but still significant. Taken from [3].

particle eigenstates. In particular, a lack of inversion symmetry of their momentum distribution around $\mathbf{k} = 0$ was observed (not shown). The single particle eigenstates on the other hand have, thanks to time reversal symmetry, an inversion symmetric momentum distribution. In the theoretical calculations, the higher energy condensate states were observed to show large variations of the phase between different high density regions. These phase variations help to build a single frequency state in a fluctuating potential landscape.

11.6 Vortices in polariton condensates

Vortices play a crucial role in the behavior of superfluids and superconductors. They are the only possibility for a superfluid to carry angular momentum and for superconductors to be pierced by a magnetic field. A vortex consists of a singularity in the order parameter phase ϕ and zero density at this singular point. Due to the single-valuedness of the order parameter, the angular momentum of superfluids is quantized, in stark contrast with classical fluids that can assume any value of angu-

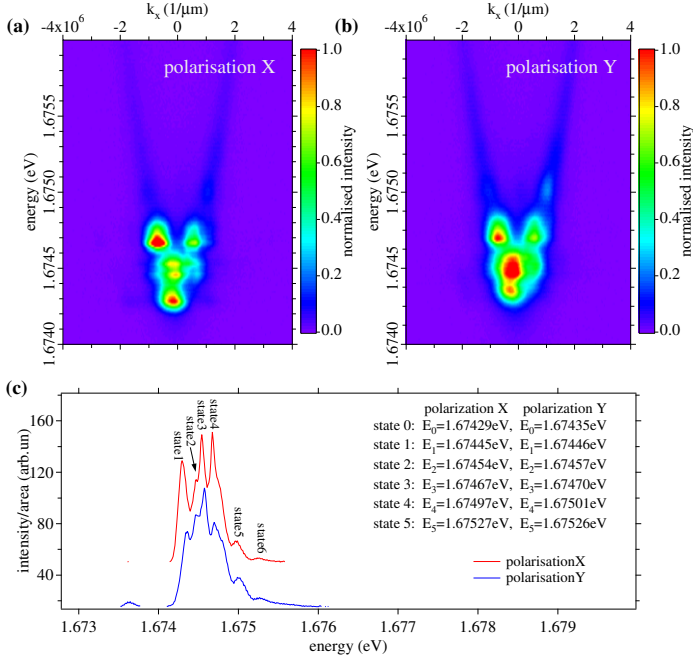


Fig. 11.6 (a),(b) photoluminescence of polaritons recorded in the (Momentum, Energy) dispersion plane and for two orthogonal polarization directions X and Y.(c) Momentum integrated photoluminescence spectra. Taken from [23].

lar momentum. The superfluid velocity associated to a quantized vortex decays as the inverse of the distance to the vortex: $\mathbf{v} = \frac{1}{r} \mathbf{1}_\phi$.

11.6.1 Quantized vortices

We have discussed above how polariton currents are generated in a finite size condensate due to spatially inhomogeneous polariton density. A natural question is whether these nonequilibrium currents could show vorticity in a disordered environment. The answer turns out to be positive. The experimental observation of quantized vortices in polariton condensates were reported in [27]. Vortices were evidenced by interferometric measurements: the characteristic phase-winding around the vortex core results in a fork-like dislocation in the interference pattern generated by overlapping the vortex area with another area of the superfluid with homogeneous phase (cf. Figs.11.8.a and 11.8.c). For their interpretation, two distinct mechanisms could be put forward. First, one could imagine the spontaneous creation of vortex-antivortex pairs, as observed in two dimensional Bose Einstein condensates with

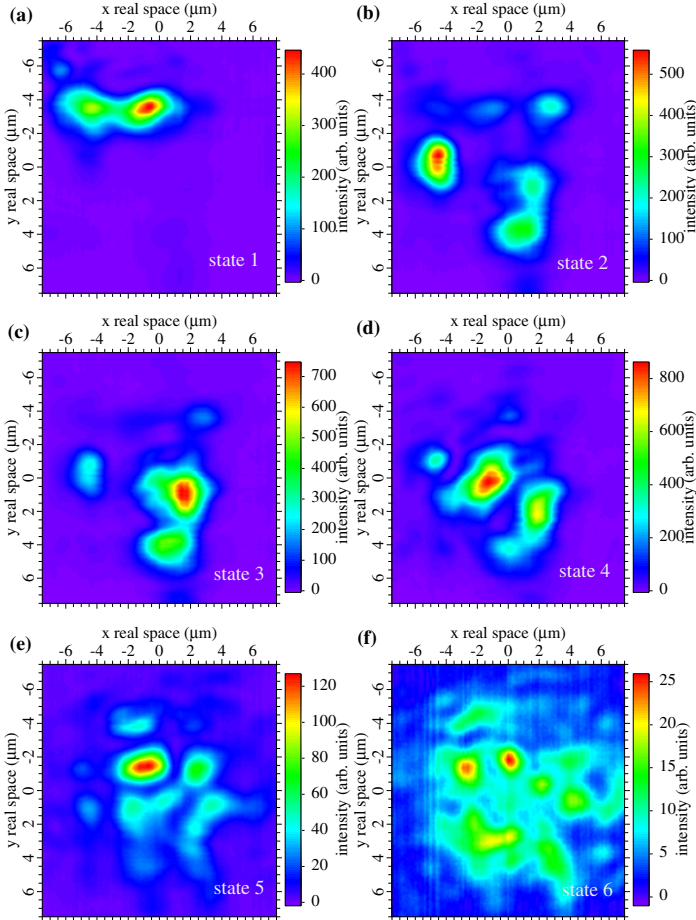


Fig. 11.7 (a)-(f): Spectrally filtered direct space photoluminescence images recorded at the energy of mode 1 (a) to mode 6 (f). Taken from [23].

ultracold atomic gases [17]. The second mechanism would be a deterministic flow within the disordered potential landscape. The first mechanism cannot hold considering the fact that measurements are integrated over a very long (seconds) time compared to any characteristic time of the condensate. Then the random fluctuations in vortex sign and position would result in washing out the fork-like pattern. On the other hand, a deterministic mechanism is supported by theoretical works: vortices are predicted in numerical simulations with the extended GP equation [27] and within the CGLE [22]. This mechanism is, to the best of our understanding, not fundamentally different from the one that leads to the formation of a polariton condensate ring in momentum space in the case of a small excitation spot (cf. sec-

tion 11.5.2). The only difference in the vortex case is that singular points in the flow pattern could be generated by some disorder potential landscapes. Because the polariton condensate extracts energy from the exciton reservoir, these vortices are in a sense not very different from vortices in a driven flow of an ordinary fluid; still the coherence of the polariton field imposes their quantization, which is a unique quantum feature.

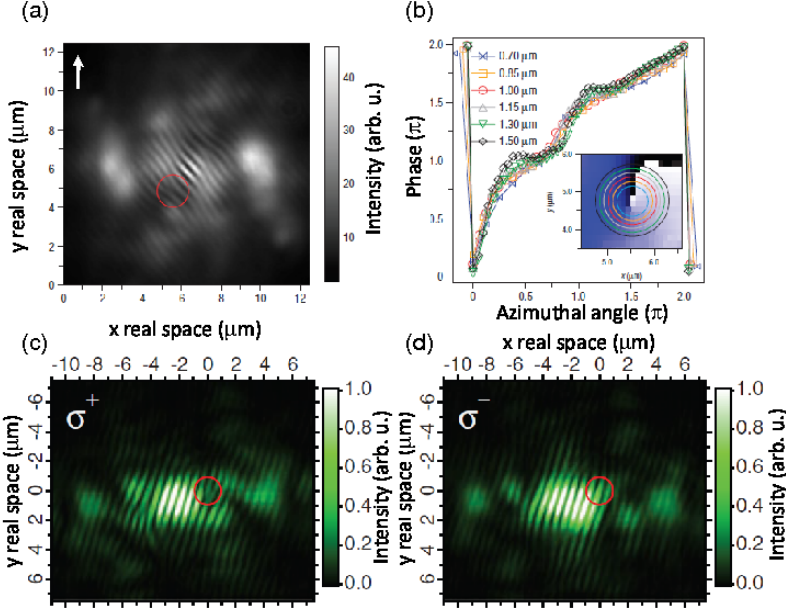


Fig. 11.8 Experimental observation of vortices in a single component polariton condensate (a) and (b) (taken from [27]) and half-quantized vortices in a two component "spinor" condensate (c) and (d) (taken from [26]). (a) Typical Interferogram due to a full vortex: the fork-like dislocation associated to the vortex can be seen inside the red circle. The condensate is strongly linearly polarized as expected. (b) Measured phase of the order parameter along the circumferences of loops centered on the vortex core and of radii ranging from $0.7\mu\text{m}$ (blue symbol) to $1.5\mu\text{m}$ (black diamonds). The inset shows a real space mapping of the order parameter phase in false colors (black for $\phi = 0$, blue for $\phi = \pi$ and white for $\phi = 2\pi$). The loops of different radii used for the main plot are also shown with different colors. (c) and (d) are typical interferograms showing evidence of a half-quantized vortices : the fork-like dislocation is observed in (c) in circular σ^+ polarization but not in the opposite σ^- polarization (d).

11.6.2 Half-quantized vortices

In an ideal planar microcavity, polaritons are not scalar particles. Due to the light-matter coupling selection rule in polarization, they have a spin-1/2-like degree of freedom, i.e. the ground state is twice degenerate. In Telluride microcavities, however, this two-fold degeneracy is lifted by a slight crystalline anisotropy [20], resulting in a singlet ground state. In this case, polaritons behave as scalar particles, condensing in the ground state with a well defined linear polarization, and featuring quantized vortices as shown in Figs.11.8.a and b.

On the other hand, in some particular places of the sample, this crystalline anisotropy is reduced by the local disorder, thus restoring the polarization degree of freedom : the polarization of the condensate can take any value at no cost in energy. Then the polariton condensate is a so-called "spinor" condensate. Vorticity of a spinor order parameter is more complicated than in the scalar case. A vortex can occur in one polarization, but not in the other one. Consider for example

$$\psi_{\uparrow}(\mathbf{r}) = f_{\uparrow}(r)e^{i\theta} \quad (11.9)$$

$$\psi_{\downarrow}(\mathbf{r}) = f_{\downarrow}(r). \quad (11.10)$$

where $\psi_{\downarrow}(\mathbf{r})$ and $\psi_{\uparrow}(\mathbf{r})$ are the order parameter two components in the circular σ^+ and σ^- polarization basis respectively. $f_{\downarrow\uparrow}(r)$ and θ are the corresponding amplitudes and relative spatial phase. In the basis of linear polarization, the two component condensate wave function takes the form

$$\psi_{\leftrightarrow}(\mathbf{r}) = e^{i\theta/2}[e^{i\theta/2}f_{\uparrow}(r) + e^{-i\theta/2}f_{\downarrow}(r)] \quad (11.11)$$

$$\psi_{\downarrow\uparrow}(\mathbf{r}) = e^{i\theta/2}[e^{i\theta/2}f_{\uparrow}(r) - e^{-i\theta/2}f_{\downarrow}(r)]. \quad (11.12)$$

If the polarization of the condensate is linear at distances far from the vortex core, we have that $f_{\uparrow} = f_{\downarrow} = f$. The order parameter then reads:

$$\psi_{\leftrightarrow}(\mathbf{r}) = e^{i\theta/2}f \cos(\theta/2) \quad (11.13)$$

$$\psi_{\downarrow\uparrow}(\mathbf{r}) = e^{i\theta/2}f \sin(\theta/2). \quad (11.14)$$

In the linear polarization basis, the condensate phase varies by π only along a closed loop around the vortex core. The continuity of the order parameter is ensured by the simultaneous rotation of the polarization. These half-quantized vortices have been recently reported in polariton condensates by K. G. Lagoudakis and co-workers [26] : interferograms have been measured as previously in both circular polarizations. A fork-like dislocation is clearly visible in σ^+ polarization as shown in Fig.11.8.c (red circle) but it is not present at the same place in the opposite polarization (Fig.11.8.d, red circle). Eq.11.14 describing a half-vortex was obtained assuming a linearly polarized condensate without preferred polarization direction. For polaritons in an isotropic environment, and in the limit of thermal equilibrium, this polarization behavior is likely to result from the interactions between cross-

circularly-polarized polaritons. The driving mechanism for the formation of half-quantized vortices is supposed to be the same as for the full vortices, with the additional ingredient that the microcavity disorder is polarization dependent and can therefore induce a rotation of the polariton polarization. Interestingly, it was shown in a recent theoretical discussion that in the case of linear polarization splitting, half vortices can still exist, but with a peculiar phase and polarization direction distribution in space : they carry a so-called "vortex string" (soliton) [68].

11.7 Correlations within a degenerate polaritons gas

11.7.1 Spatial first order correlations

In a classical gas of bosons far from the critical conditions (below the critical density or above the critical temperature), the order parameter defined as the condensate wavefunction of the form $\sqrt{N_0}\psi_0$, where N_0 is the number of particles in the condensate and ψ_0 is the zero momentum single particle wavefunction, vanishes. Upon crossing the phase boundary, it becomes significant and eventually dominates the whole system. Hence, the clear-cut demonstration of BEC is obtained by a careful measurement of the order parameter. Experimentally, the latter can be accessed using the Wiener-Khinchin identity which states that a distribution of the form $N_0\delta(\mathbf{k})$ in momentum space corresponds to infinite range first order correlations in direct space. This is the Penrose-Onsager [36] criterion, which can be investigated experimentally by interferometric measurements in direct space. Note that this criterion is also valid for a laser in a planar cavity. However, as explained before, in this case the bosonic field is of different nature. As previously mentioned, characteristics of the polariton field inside the microcavity are transmitted to the outside world thanks to the radiative recombination of polaritons at the microcavity mirror/air interface. Thus the first-order spatial correlation of the polariton system can be measured by analyzing the microcavity emission spot with an imaging Michelson interferometer, which was appropriately modified in order to extract a spatial correlation mapping of the following form:

$$g^{(1)}[(x,y),(-x,-y)] = \frac{\langle E(x,y)E^*(-x,-y) \rangle}{\langle E(x,y) \rangle \langle E^*(-x,-y) \rangle} \quad (11.15)$$

where $g^{(1)}[(x,y),(-x,-y)]$ is the normalized amount of first order correlation between both points (x,y) and $(-x,-y)$. $E(x,y)$ is the polariton photoluminescence at the specified position, and the averaging $\langle \rangle$ is performed over time. A typical result is shown in Fig.11.9 for a polariton gas of $25\mu m$ diameter, i.e. much larger than the de Broglie wavelength $1\mu m$. The point $(x=0, y=0)$ of the autocorrelation is chosen in the middle of the polariton gas.

Below threshold, a very short range correlation peak of about $1\mu m$ diameter is observed (Fig.11.9.a). For a polariton gas in thermal equilibrium (like just be-

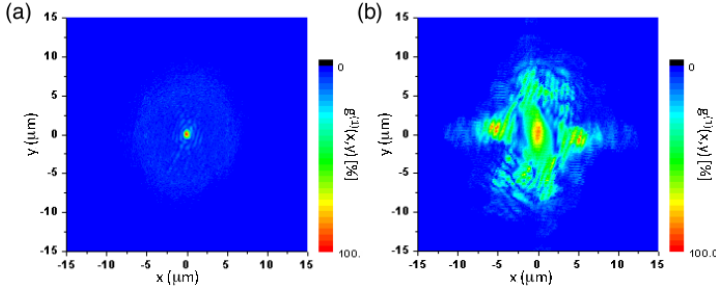


Fig. 11.9 Correlation mapping of the polariton gas below (a) and above (b) BEC threshold. The color scale represents the normalized first-order space correlation $g(1)$ between the points (x,y) and $(-x,-y)$. The autocorrelation point $(0,0)$ of the image has been chosen experimentally in the middle of the condensate. Taken from [20]

fore threshold) this correlation length would correspond to the polariton thermal de Broglie wavelength. Above threshold (Fig.11.9.b) long range correlations appear, limited only by the $25\mu\text{m}$ diameter of the condensate. This demonstrates unambiguously that the Penrose-Onsager criterion for BEC is satisfied. The inhomogeneous character of correlation in the condensed regime directly reveals the inhomogeneous density distribution of the polariton condensate resulting from disorder.

A quantitative understanding of the observed spatial coherence in polariton condensation experiments remains a challenging issue, mainly because the disorder strongly modulates the spatial coherence. At thermal equilibrium and in a disorderless system, a power law decay of the coherence is expected for a Bose gas in the superfluid phase. Several theoretical studies have highlighted the fact that the nonequilibrium situation reduces the spatial coherence. At the level of the Boltzmann equation, this can be understood as a consequence of the bottleneck effect, which is a purely nonequilibrium feature, and which favors the occupation of the excited states instead of the lower energy ones. The spatial coherence being the Fourier transform of the momentum distribution, a larger occupation of the excited states implies a reduced spatial coherence.

By building a kinetic theory for the Bogoliubov excitations of the condensate which includes the coherence, it was shown by Sarchi and Savona that anomalous scattering (scattering within the condensate toward excited states) also contributes to deplete the condensate and decrease the spatial coherence [66].

11.7.1.1 Comparison with the spatial first order correlations in a VCSEL

Semiconductor microcavity lasers in the weak coupling regime (VCSEL) have in principle the same potential to exhibit long range spatial coherence. In practice, however, the observed spatial coherence is often limited to a few microns, much smaller than the size of the emitting area [32, 62]. The interpretation of these obser-

vations is that the spatial mode selection is not sufficiently efficient in order to select a single macroscopically occupied mode. Instead, the light field forms filaments.

The elucidation of the difference between the polariton condensation regime and weak coupling lasing regime requires further study, but it is likely that the energy dependence of the gain plays an important role. The relaxation from the exciton reservoir to the lower polariton branch favors the occupation of the lowest energy states, whereas in the weak coupling lasing case a weaker energy dependence of the relaxation is expected. Numerical simulations with a Wigner representation of the Bose field have indeed shown that the energy dependence of the relaxation is crucial to obtain long range spatial coherence [63]. The spatial coherence in the VCSEL regime can be enhanced by patterning a microcavity with a metal grid (that introduces spatially selective losses). It turns out that mode selection is more effective and that much longer coherence lengths are obtained. It was demonstrated by Lundeberg et al. [61] that a VCSEL array can exhibit very good spatial coherence up to 36 microns (the full system size).

11.7.1.2 First order time correlations

The coherence time of polariton condensates was assessed experimentally as well [30]. Below threshold, the coherence time is fixed by the cavity life time which amounts to a few ps. Above threshold, the coherence time is enhanced by two orders of magnitude and reaches hundreds of ps. The coherence decay was exponential at all densities. In the infinite 2D Bose gas, one expects a power law decay of the coherence, but if the condensate size is too small to accommodate many spatial excitations, an exponential decay of temporal correlations is predicted [64]. A detailed discussion on this point is presented chapter 12 section 6. Contrary to the Schawlow-Townes increase of the coherence time with density (in the case of lasers), it was observed to saturate and decrease at high densities. This behavior was explained as an effect of polariton interactions [30, 65]. In the presence of interactions, density fluctuations cause a fluctuating blue shift, which in turn results in a reduced phase coherence.

11.7.2 Number fluctuations in a polariton condensate

Second-order time correlation characterizes the particle number fluctuations within a many-particle system, providing crucial information on its state. For instance it permits to distinguish between two spectrally narrow light sources of different nature, like a laser and an excited atom gas emitting photoluminescence. Although both sources share similar time coherence properties (within the first order), the former exhibits the lowest possible classical fluctuations also called "shot noise" limit (quantitatively, it yields $g^{(2)}(0) = 1$). The latter shows larger fluctuations ($g^{(2)}(0) = 2$) of thermal origin. A Bose-Einstein condensate is very similar to a laser field and presents a similar behavior in terms of particle number fluctuations: this

fact has been nicely verified in the case of cold atom condensate [51]. As explained previously the polariton number time fluctuations is reflected directly on the emission intensity fluctuations. Hence, the measurement can be done using a Hanbury Brown-Twiss interferometer [7]. Such measurements have been performed independently in two different groups with identical Telluride-based microcavities [30, 19]. In the first reference, the use of a low noise laser (diode laser as in section 11.5.3.1) permitted better resolved measurements below and at the critical density. The second reference provides data in a higher density regime. In the low polariton density limit, the polariton field is incoherent and the fluctuations should exceed significantly the shot noise limit. In practice however, it is very difficult to obtain a quantitative measurement because the coherence time, which fixes the timescale of the fluctuations, is of $\sim 1.5ps$, i.e. 50 times shorter than the instrumental resolution (limitation in the case of CW excitation measurement) and 100 times shorter than the free e-h to polariton relaxation time (limitation in the case of a pulsed excitation measurement). Nevertheless, with an integration time long enough, above shot-noise fluctuations (i.e. photon bunching) could be clearly observed in both reports. A measurement is shown in Fig.11.10.a : the observed small deviation from $g^{(2)}(0) = 1$ actually corresponds to a much larger deviation which is difficult to evaluate quantitatively, due to the low instrumental resolution as explained above.

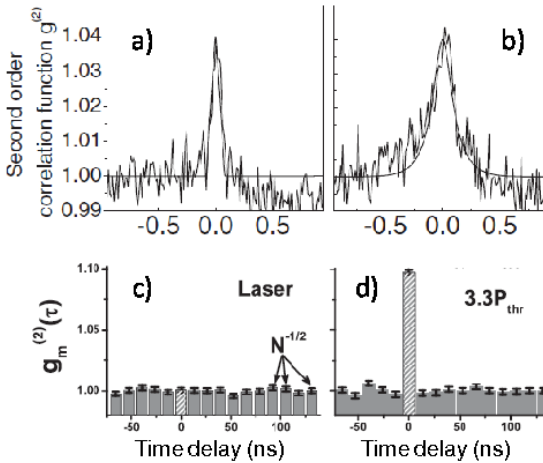


Fig. 11.10 Measured second order time correlation function $g^2(\tau)$ of the polariton condensate below (a) and at (b) the critical density. The optical excitation is provided by a semiconductor laser. Taken from [30]. (d) Measured $g^2(\tau)$ of the polariton condensate at three time the critical density. The excitation is provided by picosecond pulses from a Ti:Sapphire laser. Taken from [19]. (c) Measured $g^2(\tau)$ of the Ti:Sapphire laser used for comparison with (d).

Upon crossing the phase boundary, the fluctuations are found to drop to a value close to the shot noise limit, as expected for a coherent state. This is shown in

Fig.11.10.b. As opposed to the low density case, in this regime the measurement provides directly the right value of $g^{(2)}(0)$ (Fig.11.10.b) thanks to the strongly enhanced coherence time of $\sim 150ps$ of the polariton condensate. Interestingly, upon increasing further the polariton density, fluctuation increases again, as a result of increasing interactions between polaritons and the environment (other polaritons, excitons and free carriers). This is shown in Fig.11.10.d in the pulsed excitation regime. To make sure these strong fluctuations are not due to the excitation, the laser fluctuations are shown in Fig.11.10.c and found to be shot noise limited. This fluctuation increase is consistent with the observed linewidth broadening of the condensate spectrum far above threshold (not shown). Conventional photon lasers exhibit an opposite behavior : for a gas laser like He-Ne, the fluctuations are found to drop to the shot noise limit upon crossing the threshold and remain there even ten times above threshold [9]. In a VCSEL, the behavior of the photon number fluctuations has not been measured yet to the best of our knowledge.

Second order time correlations were measured in 2002 in an Arsenide-based microcavity where the polariton gas was fed by resonant injection of cold excitons (instead of hot free e-h pairs in the case of non-resonant excitation). Fluctuations measured across the phase boundary showed a somewhat opposite behavior with respect to the above description [13]: they were found to steadily drop from a high value at threshold to a lower value at densities exceeding ten times the critical density. This behavior is not clearly understood at present.

11.8 Conclusion and Outlook

In this short review we have highlighted some of the unique properties of polaritons as a Bose gas. We have seen how experiments can be properly explained by theoretical descriptions requiring an interesting mixture of equilibrium Bose gas and laser theories. Progress on the physics of microcavity excitonic-polaritons are intimately connected with technological developments in semiconductor nanofabrication. In 1992, the strong coupling regime was achieved for the first time in a semiconductor device thanks to new technological advances in the domain of semiconductor epitaxy of Arsenide alloys [53]. Five years later, more robust polariton gases could be obtained thanks to the epitaxy development of Telluride alloys [2], which permitted nine years later the realization of the first polariton BEC [20].

In present days, in order to gain new insights into polariton Bose gases and to explore practical applications, new photonic devices and materials are developed to provide a better control of the polariton environment. Recently, Arsenide microcavities have reached quality factors high enough to lower the critical density to achieve Bose quantum degeneracy [54]. With the multiple possibilities in terms of nanofabrication, Arsenide-based devices could become a powerful playground for polariton condensate in the dilute and low temperature limit [5]. Nanofabrication techniques of wider bandgap materials (GaN, ZnSe and ZnO) are also quickly evolving. With these materials, the polaritons are intrinsically more robust and po-

tentially capable of showing spontaneous coherence at room temperature and very high density, deep into the condensed phase. Thus, planar microcavities of high quality factors with bulk or quantum well active medium are currently fabricated with Nitride [10] and zinc oxide-based materials [33]. Selenide-based materials are also very promising, where high quality microcavities and advanced etching techniques are available [24]. On the other hand, different ways of confining the light to obtain the strong coupling regime are currently under study, like zinc oxide microwires [47] and Nitride-based microdisks [45, 48]. In the former, 1D polaritons have been observed at room temperature, with negligible thermal broadening and unprecedented figure-of-merit (Rabi splitting/polariton FWHM) > 70 [67].

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Chapter 12

Keldysh Green's function approach to coherence in a non-equilibrium steady state: connecting Bose-Einstein condensation and lasing

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Abstract Solid state quantum condensates often differ from previous examples of condensates (such as Helium, ultra-cold atomic gases, and superconductors) in that the quasiparticles condensing have relatively short lifetimes, and so as for lasers, external pumping is required to maintain a steady state. On the other hand, compared to lasers, the quasiparticles are generally more strongly interacting, and therefore better able to thermalise. This leads to questions of how to describe such non-equilibrium condensates, and their relation to equilibrium condensates and lasers. This chapter discusses in detail how the non-equilibrium Green's function approach can be applied to the description of such a non-equilibrium condensate, in particular, a system of microcavity polaritons, driven out of equilibrium by coupling to multiple baths. By considering the steady states, and fluctuations about them, it is possible to provide a description that relates both to equilibrium condensation and to lasing, while at the same time, making clear the differences from simple lasers.

12.1 Introduction

Bose-Einstein condensation (BEC), the equilibrium phase transition of weakly interacting bosons, was realised over ten years ago in ultra-cold atomic gases. After

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long and strenuous efforts to observe this state in solids, BEC of polaritons[1] and of magnons[2] were reported. These reports followed observations of related effects for excitons in quantum Hall bilayers[3], spin triplet states in magnetic insulators[4] and excitons in coupled quantum wells[5, 6, 7]. Solid-state condensates depart from the archetypal BEC in several ways. Most importantly they live for short times and rely on external pumping. Indeed, it was the decay, and consequent lack of equilibrium, which for a long time presented the obstacle the realisation of solid-state BEC. Even if one can accelerate thermalisation, the decay, and the consequent flux of particles, remains a more important effect in solid state than it generally does in cold atomic gases, or in other quantum condensates such as superfluid Helium.

When considering whether such a system may be treated as equilibrium or not, there are several distinct characterisations of the degree to which the system is non-equilibrium. The most obvious compares particle lifetime to the time required for collisions to thermalise the system, determining the extent to which a thermal distribution may arise. The timescale for establishing a thermal distribution within one part of the system can however be quite different to that for establishing either thermal or chemical equilibrium between different parts of the system. Another characterisation of whether non-equilibrium physics is relevant arises from comparing the linewidth due to finite particle lifetime to the temperature of the system, thus determining whether lifetime or temperature effects dominate coherence properties. Table 12.1 summarises the typical timescales and energy scales connected with different examples of metastable quantum condensates. It is clear that the ratio of thermalisation time to the particle lifetime is generally somewhat larger for solid-state condensates than it is for cold atomic gases. If one instead compares the ratio of the linewidth due to decay to the characteristic temperature, polaritons stand out as having a decay linewidth of the same order of magnitude as their temperature. As such, polaritons are good systems in which to study effects of finite lifetime on coherence properties.

Table 12.1 Characteristic timescales and energies for: particle lifetimes, times to establish a thermal distribution, linewidth due to finite lifetime, and characteristic temperatures for various candidate condensates. Comparison of the first two describes how thermal the distribution will be; comparison of the later two determine the effect of finite lifetime on coherence properties.

	Lifetime Thermalisation		Linewidth	Temperature	
Atoms[8]	10s	10ms	2.5×10^{-13} meV	10^{-8} K	10^{-9} meV
Excitons[9]	50ns	0.2ns	5×10^{-5} meV	1K	0.1meV
Polaritons[10]	5ps	0.5ps	0.5meV	20K	2meV
Magnons[2]	1 μ s	100ns	2.5×10^{-6} meV	300K	30meV

Because, as we will discuss further below, polariton condensates provide such a clear illustration of the properties of non-equilibrium condensation, we will focus on them in particular. Microcavity polaritons are the quasiparticles which result from strong coupling between photons confined in a semiconductor microcavity, and excitons in a quantum well. By changing the detuning between the excitons and photons, and by changing the strength of an external pump that injects

polaritons, one can modify the polariton mass, density and the effect of interactions between polaritons. A more detailed introduction to microcavity polaritons and semiconductor microcavities can be found in several review articles and books [11, 12, 13, 14, 15, 16, 17] (see also Chapter 11).

The intrinsic non-equilibrium and dissipative nature of solid-state condensates, especially of polaritons, brings connections to other systems exhibiting macroscopic coherence, i.e. lasers. With the realisation of more complex, interaction dominated lasers, such as random lasers (see e.g. Refs. [18, 19]) or atom lasers (e.g. Refs. [20, 21, 22]), this connection is particularly pronounced. Compared to simple lasers, polaritons are however more strongly interacting, and therefore much better able to thermalise than are photons, and so in many ways solid-state condensates can be viewed as being somewhere in between an equilibrium BEC and a laser. At the same time, at large temperatures and/or in the presence of large decoherence mechanisms and large pumping the same microcavity system supports a simple lasing action. In this context, microcavity polaritons provide particularly excellent playground for studying coherence in a dissipative environment, and the differences and similarities between condensates and lasers. Clearly, an approach which takes into account the non-equilibrium and dissipative nature of this new state of matter, as well as strong interactions, multimode-structure, low dimensionality and finite size is necessary.

This chapter will discuss a theoretical approach to modelling quantum condensates that are driven out of equilibrium by a flow of particles through the system. We therefore consider coupling the system to baths, which can transfer energy as well as particles to and from the system. With such baths, we find that the behaviour of a simple laser can be recovered in the limit of high temperature baths. A different scenario of how decoherence affects condensation can be found if one considers static disorder — i.e. allowing scattering, but with no transfer of energy to or from the system. Such a problem [23, 24] is closely related to the Abrikosov-Gorkov approach to disordered superconductors[25]. As in the case of superconductors, one finds a distinction between “pair-breaking” and “non-pair-breaking” disorder (respectively magnetic and non-magnetic impurities in the superconducting case). As expected from Anderson’s theorem[26], the coherence associated with the condensate leads to a gap in the exciton density of states, which makes the condensate robust to non-pair-breaking disorder. With pair-breaking disorder, decoherence eventually destroys the gap and finally the condensate, but for small amounts of decoherence, the gap protects the condensate. A similar scenario also exists in the ultra high density limit, where excitons are destroyed by screening, leading to an electron-hole plasma phase[27], which can nonetheless support lasing. While we focus in this chapter instead on the effects of particle flux, and baths that can transfer energy, these other results illustrate that there are a variety of ways in which decoherence can either suppress or modify the properties of a condensate. In principle one can have both a crossover from a polariton condensate to a regular laser (weak coupling but still excitonic gain medium, as discussed here), and a crossover to a particle-hole laser (weak coupling, electron-hole plasma, if screening is strong).

The approach to modelling the condensate with a flux of particles presented in this chapter is based on work by the authors in Refs.[28, 29]. While in those works,

the results were derived and presented making use of the non-equilibrium path integral approach[30], both the results and their theoretical basis can be understood without this technical background, by considering the diagrammatic approach to calculating non-equilibrium Green's functions [31, 32, 33]. The particular aim of this chapter is therefore to review some of these results, illustrating in some detail how a steady state non-equilibrium system which develops spontaneous coherence, can be treated in the non-equilibrium diagrammatic formalism. At the same time, this approach will provide a natural language to highlight the way this system relates both to equilibrium condensates and to lasers, and to understand the ingredients that makes it differ from these limits.

There are a number of other known approaches to describing systems driven out of equilibrium by coupling to multiple baths. Those that have been applied to microcavity polaritons include: quantum kinetic equations[34, 35, 36, 37, 38, 39, 40, 41], Heisenberg-Langevin equations[42], stochastic methods for density matrix evolution (i.e. truncated Wigner approximation) [43, 44]; as well as mean-field approaches, considering the complex Gross-Pitaevskii equation, in some cases including also coupling to reservoirs or thermal baths [45, 46, 47] (see Chapter 11 for more details). While this chapter does not intend to review the merits of each of these approaches, it is worth noting that in general, these approaches are all connected. The connections between many of them can simply be seen by looking at their relation to the non-equilibrium diagrammatic approach. As discussed in [33, 48], the quantum Boltzmann equation can be derived as an equation for the distribution function that appears in the Keldysh Green's function, along with a Wigner transformation from $F(t, r, t', r')$ to $F(T, R, \omega, p)$. It will become clear from the discussion in Sec. 12.4.1, that there is a close analogy between the Keldysh Green's functions and the Heisenberg-Langevin equations, with the bath Green's functions describing the same physics as the correlation functions of the bath noise operators in the Heisenberg-Langevin approach. There also exists a connection between the approach described here and density matrix evolution. The single particle density matrix is given by $\langle \psi^\dagger(r, t) \psi(r', t) \rangle$, and thus corresponds to an appropriate combination of equal time Green's functions. The density matrix naturally gives single time expectations of appropriate observables, it is also possible to derive two-time correlations from the time evolution of the density matrix, by means of the quantum regression theorem[49]. The quantum regression theorem however relies on making an additional Markov approximation regarding the bath occupations, as well as a Markov approximation for the bath density of states[50]. The Keldysh Green's function approach does not require this additional Markov approximation; and in fact Sec. 12.5.2.1 will show how making this further approximation restricts the conditions for condensation to occur. In order to illustrate the application of the non-equilibrium technique, we consider a specific model of microcavity polaritons, starting from disorder localised excitons strongly coupled to cavity photons[51, 52, 53]. In this model, interactions between excitons are included by treating the excitons as hard-core bosons, allowing one exciton, but no more, to occupy a given disorder localised state. For the discussion presented here, using this model provides a number of technical advantages: it connects closely to the idea of gain from two-level sys-

tems that is typically used in models of simple lasers[49], making the comparison to lasing straightforward; and it automatically includes nonlinearity of the excitons, allowing this nonlinearity to be described by the properties of the exciton representation, rather than requiring higher order diagrammatic corrections. In addition, in an equilibrium situation, the mean-field theory of this model is known to give a reasonable description of the critical temperature, except at very low densities where fluctuation corrections become important[52].

This chapter is organised as follows; section 12.2 introduces the model Hamiltonian, and its coupling to baths. Section 12.3 then describes the approach we will take to modelling this system, reviewing some standard results of the non-equilibrium diagrammatic technique that will be used later, and discussing the mean-field approach we use to find the steady state. In order to evaluate this mean-field condition, it is necessary to determine the effects of the baths on the system, by calculating particular self energy diagrams, these self energies are presented in section 12.4. Section 12.5 then discusses the mean-field theory, considering how it can recover both equilibrium results in one limit, as well as the description of a simple laser in another limit. Section 12.6 discusses fluctuations about the steady state, analysing stability, and further illuminating the connection to (and distinctions from) a simple laser; section 12.7 then provides a more qualitative discussion of the fluctuations of the condensed system, focusing in particular on the combined effect of finite size and finite lifetimes.

12.2 Polariton system Hamiltonian, and coupling to baths

As explained above, we consider a model of excitons as hard core bosons coupled to propagating photons. To write the Hamiltonian for hard-core bosons, it is convenient to introduce fermionic operators $b_i^\dagger, a_i^\dagger$, such that the two fermionic states represent the presence or absence of an exciton on a given site, hence the operator $b_i^\dagger a_i$ is the exciton creation operator. With this notation, the system is described by:

$$H_{\text{sys}} = \sum_i \varepsilon_i (b_i^\dagger b_i - a_i^\dagger a_i) + \sum_k \omega_k \psi_k^\dagger \psi_k + \sum_{i,k} g_i (\psi_k^\dagger a_i^\dagger b_i + \text{H.c.}), \quad (12.1)$$

where ε_i is the energy of a localised exciton state, g_i is the exciton-photon coupling strength, and $\omega_k = \omega_0 + k^2/2m_{\text{phot}}$ is the dispersion of cavity photons.

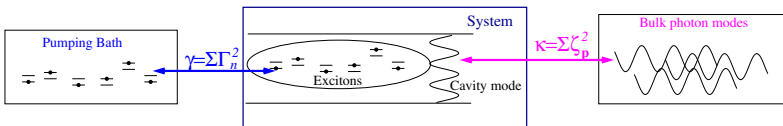


Fig. 12.1 Cartoon of system, consisting of photons strongly coupled to excitons, and external baths, describing pumping and decay. Adapted from Ref.[29].

As sketched in Fig. 12.1, this can then be driven out of equilibrium by coupling to two baths, so the the system evolves under the full Hamiltonian $H = H_{\text{sys}} + H_{\text{sys,bath}} + H_{\text{bath}}$. It will be useful later on to divide the coupling to baths into coupling to the pumping bath, and coupling to the decay bath $H_{\text{sys,bath}} = H_{\text{sys,bath}}^{\text{pump}} + H_{\text{sys,bath}}^{\text{decay}}$ where the forms of the coupling to the pumping and decay baths are:

$$H_{\text{sys,bath}}^{\text{pump}} = \sum_{n,i} \Gamma_{n,i} \left(a_i^\dagger A_n + b_i^\dagger B_n + \text{H.c.} \right), \quad H_{\text{sys,bath}}^{\text{decay}} = \sum_{p,k} \zeta_{p,k} \left(\psi_k^\dagger \Psi_p + \text{H.c.} \right). \quad (12.2)$$

Here $\Gamma_{n,i}$ is the coupling to a pumping bath, described by the fermionic operators $B_n^\dagger, A_n^\dagger$, and $\zeta_{p,k}$ is the coupling to decay bath, describing bulk photon modes $\Psi_p^\dagger$.

The bath Hamiltonian is taken to have the simple quadratic form:

$$H_{\text{bath}} = \sum_n v_n^\Gamma (B_n^\dagger B_n - A_n^\dagger A_n) + \sum_p \omega_p^\zeta \Psi_p^\dagger \Psi_p \quad (12.3)$$

Describing the pumping reservoir and photon decay as baths means that we assume these both contain many modes (i.e. are much larger than the system), and thermalise rapidly compared to the interaction with the system. These assumptions mean that one may impose a particular distribution function on the bath modes, and then determine what distribution the system adopts; we will take a thermal distribution for the pumping bath, specified by a bath temperature and chemical potential, and we will assume the bulk photon modes are unoccupied. Note that we do not explicitly introduce any system chemical potential, as the density of the system will be fixed by the balance of pumping and decay, however a natural definition of the system chemical potential will arise later.

12.3 Modelling the non-equilibrium system

The Keldysh non-equilibrium diagrammatic technique[31] is an approach well suited to dealing with the kind of non-equilibrium steady state which we consider here. Section 12.3.1 briefly summarises the concepts that will be important in the remainder of this chapter; for a more complete introduction, see for example Refs. [31, 32, 33]. Within this diagrammatic approach, we will then determine the possible steady states of the system by a mean-field approach, introduced in section 12.3.2.

12.3.1 Non-equilibrium diagram approach

In order to determine both the spectrum (i.e. the ground and excited states, taking into account interactions and coupling to baths), and the non-equilibrium occupation of this spectrum, it is necessary to calculate two linearly independent Green's functions; it is convenient to make these the retarded and Keldysh Green's functions:

$$D^R(t, r) = -i\theta(t) \langle [\psi(t, r), \psi^\dagger(0, 0)]_- \rangle, \quad D^K(t, r) = -i \langle [\psi(t, r), \psi^\dagger(0, 0)]_+ \rangle. \quad (12.4)$$

Here, $[\psi, \psi^\dagger]_\mp$ indicates the commutator (anti-commutator) of ψ and $\psi^\dagger$. These Green's functions can be written as time-ordered products of fields by introducing the Keldysh contour, shown in Fig. 12.2. Each point on this contour is labelled by

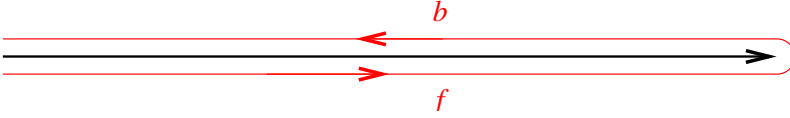


Fig. 12.2 Keldysh closed-time-path contour, which can generate multiple orderings of fields

$(t, \{f, b\})$, where the f, b label whether it is on the forward or backward branch. We then introduce the contour time ordering T_c , such that fields on the backward contour are always later than those on the forward contour, and that pairs of fields on the backward contour should appear in reverse order. By then introducing symmetric and anti-symmetric combinations of these fields $\psi_\pm = [\psi(t, f) \pm \psi(t, b)] / \sqrt{2}$, one may write the Green's function:

$$D = \begin{pmatrix} D^K & D^R \\ D^A & 0 \end{pmatrix} = -i \left\langle T_c \begin{pmatrix} \psi_+(t, r) \\ \psi_-(t, r) \end{pmatrix} \begin{pmatrix} \psi_+^\dagger(0, f), \psi_-^\dagger(0, b) \end{pmatrix} \right\rangle. \quad (12.5)$$

Here, D^A refers to the advanced Green's function, which is the Hermitian conjugate of the retarded Green's function.

Given the above time-ordered products, one may use standard methods[54, 55] to write a diagrammatic expansion, by writing the Heisenberg picture fields in terms of the interaction picture fields $\tilde{\psi}(t)$:

$$\psi(t) = U^{-1}(t) \tilde{\psi}(t) U(t), \quad \tilde{\psi}(t) = e^{iH_0 t} \psi e^{-iH_0 t}, \quad (12.6)$$

where $H = H_0 + H_{\text{int}}$, and H_0 is “free”, meaning that it is simple to write expectations of products of fields evolving according to H_0 . By formally solving the equation for $U(t)$, one may then write the Green's functions in the following form:

$$D = -i \left\langle T_c \left[\begin{pmatrix} \tilde{\psi}_+(t, r) \\ \tilde{\psi}_-(t, r) \end{pmatrix} \begin{pmatrix} \tilde{\psi}_+^\dagger(0, 0) \\ \tilde{\psi}_-^\dagger(0, 0) \end{pmatrix} U \right] \right\rangle \quad (12.7)$$

$$U = \exp \left[-i \int_C \tilde{H}_{\text{int}}(t) dt \right] = \exp \left[-i \int_{-\infty}^{\infty} (\tilde{H}_{\text{int}}(t, f) - \tilde{H}_{\text{int}}(t, b)) dt \right]. \quad (12.8)$$

The diagrammatic expansion then follows by expanding the exponential, which produces vertices coupling free fields, and connecting these vertices by lines representing the Green's functions of the free fields. Compared to other diagrammatic expansions, the only extra complication is to keep track of the $\pm$ labels on the fields, both in the matrix structure of Keldysh/retarded/advanced Green's functions, and in the form of U .

In the following, we will frequently make use of the Dyson equation[55, 54, 33], $D^{-1} = D_0^{-1} - \Sigma$, and so it is useful to record the free inverse Green's function. The inverse Green's function has the structure:

$$D^{-1} = \left[\begin{pmatrix} D^K & D^R \\ D^A & 0 \end{pmatrix} \right]^{-1} = \begin{pmatrix} 0 & [D^A]^{-1} \\ [D^R]^{-1} & [D^{-1}]^K \end{pmatrix}, \quad (12.9)$$

where $[D^{-1}]^K = -[D^R]^{-1} D^K [D^A]^{-1}$. Using the results for a free field, one has

$$[D_0^R]^{-1} = \omega - \omega_k + i\eta, \quad [D_0^{-1}]^K = (2i\eta)(2n_B(\omega) + 1), \quad (12.10)$$

where η is infinitesimal. All of the results noted above assume bosonic fields; the results for fermionic fields are similar, but commutators and anti-commutators are interchanged in the definitions of Keldysh and retarded Green's functions. For our particular model of microcavity polaritons, the division of the full Hamiltonian into H_0 and H_{int} will be to take:

$$H_0 = \sum_i \varepsilon_i (b_i^\dagger b_i - a_i^\dagger a_i) + \sum_k \omega_k \psi_k^\dagger \psi_k + \sum_i g_i \psi_0 \left(e^{i\mu_S t} a_i^\dagger b_i + e^{-i\mu_S t} b_i^\dagger a_i \right) + H_{\text{bath}} \quad (12.11)$$

where ψ_0 is a mean-field coherent photon field, as discussed in the next section. This means that H_{int} will contain the system–bath interactions, as well as the interaction between the two-level systems and incoherent photon fluctuations. In the following we will however generally focus on one part of H_{int} at a time.

12.3.2 Mean-field condition for coherent state

For a system coupled to multiple baths, the mean-field theory can no longer be thought of as minimising free energy, but rather as a stable self consistent steady state. For a condensed solution, one looks for a steady state of the form $\langle \psi_k \rangle = \psi_0 \exp(-i\mu_S t) \delta_{k,0} = \psi_0(t) \delta_{k,0}$, where μ_S is introduced here merely as part of the steady state ansatz, but it will be seen to play a role analogous to the equilib-

rium chemical potential. To be a self-consistent solution, this ansatz must satisfy the Heisenberg equation: $\langle i\partial_t \psi \rangle = \langle [\psi, H] \rangle$, and so:

$$\mu_S \psi_0(t) = \omega_0 \psi_0(t) + \sum_i g_i \langle a_i^\dagger(t) b_i(t) \rangle + \sum_p \zeta_{p,0} \langle \Psi_p(t) \rangle. \quad (12.12)$$

The expression $\langle a_i^\dagger(t) b_i(t) \rangle$ describes the polarisation of the two-level systems, and can be written in terms of the Keldysh Green's function, as:

$$\langle a_i^\dagger(t) b_i(t) \rangle = \frac{1}{2} \left\langle \left[b_i(t), a_i^\dagger(t) \right]_- \right\rangle = \frac{i}{2} G_{a_i^\dagger b_i}^K(t, t) = \frac{i}{2} \int \frac{dv}{2\pi} G_{a_i^\dagger b_i}^K(v). \quad (12.13)$$

As well as this self-consistency condition to determine the coherent field amplitude and the effective system chemical potential μ_S , the mean-field approach can also be used to give an estimate of the polariton density. This density will be used later in producing the phase diagram of the polariton condensate. The mean-field estimate of the total density is given by the combination of the photon density $|\psi_0|^2$, and the fermion density $(i/2)\text{Tr}[G_{b_i^\dagger b_i}^K - G_{a_i^\dagger a_i}^K]$.

12.4 Effects of baths on system correlation functions

In the above, we found that the mean-field condition could be written in terms of the two-level system Green's function, and the expectation of the decay bath fields. In this section we will discuss in detail the treatment of the baths and their effect on system's correlation functions, which will then determine the conditions under which a condensed solution may exist. Most of the effort, in Sec. 12.4.2, will be dedicated to finding $G_{a_i^\dagger b_i}^K(v)$ including the effects of pumping. Before doing this, section 12.4.1 will address the simpler problem of how $\langle \Psi_p(t) \rangle$ can be related to the decay bath Green's function and thus evaluated.

12.4.1 Decay bath and $\langle \Psi_p \rangle$

To calculate $\langle \Psi_p(t) \rangle$ in terms of non-equilibrium Green's functions, one may first use the interaction picture, in terms of the system-bath coupling, to write $\Psi_p(t) = U^{-1}(t) \tilde{\Psi}_p(t) U(t)$. Here, $U(t)$ is the time-ordered exponential as in Eq. (12.6):

$$U(t) = T \exp \left[-i \int_{-\infty}^t dt' \tilde{H}_{\text{sys,bath}}^{\text{decay}}(t') \right]. \quad (12.14)$$

Then, consider inserting a factor:

$$1 = T \exp \left[i \int_t^\infty dt' \tilde{H}_{\text{sys,bath}}^{\text{decay}}(t') \right] \cdot T \exp \left[-i \int_t^\infty dt' \tilde{H}_{\text{sys,bath}}^{\text{decay}}(t') \right], \quad (12.15)$$

either before or after $\tilde{\Psi}_p(t)$. The resulting expression implies that one has:

$$\langle \Psi_p(t) \rangle = \langle T_C[\tilde{\Psi}_p(t, f)U] \rangle = \langle T_C[\tilde{\Psi}_p(t, b)U] \rangle = \frac{1}{\sqrt{2}} \langle T_C[\tilde{\Psi}_{p,+}(t)U] \rangle, \quad (12.16)$$

where the last equality has made use of the fact that if the expectation of $\tilde{\Psi}(t, f)$ and $\tilde{\Psi}(t, b)$ match, then the expectation of $\tilde{\Psi}_-(t)$ must vanish. We are interested in particular in the value of this expectation $\langle \Psi_p \rangle$ when we consider the system in the mean-field approximation. In this case the system bath interaction term is given by:

$$\int_C dt \tilde{H}_{\text{sys,bath}}^{\text{decay}}(t) = \int_{-\infty}^\infty dt \sum_p \zeta_{p,0} \sqrt{2} [\tilde{\Psi}_{p,-}^\dagger(t) \psi_0(t) + \psi_0^*(t) \tilde{\Psi}_{p,-}(t)]. \quad (12.17)$$

With vertices given by this interaction, the set of diagrams involved in evaluating Eq. (12.16) is particularly simple: the only possible connected diagram is one with a single bath Green's function connecting the source term in $H_{\text{sys,bath}}$ to the field $\tilde{\Psi}_+$ that we want to measure. As such, the sum appearing in Eq. (12.12) can be written as:

$$\sum_p \zeta_{p,0} \langle \Psi_p(t) \rangle = \sum_p \zeta_{p,0}^2 \int dt' D_{\Psi_p^\dagger \Psi_p}^R(t, t') \psi_0(t'). \quad (12.18)$$

The simple form this equation takes is also the form one would find by making the Born approximation; i.e. assuming that $\zeta_{p,0}$ is small, so that terms like $\sum_p \zeta_{p,0}^2$ should be kept, and neglecting terms involving any higher power of $\zeta_{p,0}$. However, in the current case, because of the linearity of the coupling, no other connected diagrams exist, so no assumption of smallness is required in order to neglect higher order terms.

For a free bath, one may write $\tilde{\Psi}_p(t) = e^{-i\omega_p^\zeta t} \Psi_p$, and so the bath Green's function is given by $D_{\Psi_p^\dagger \Psi_p}^R(t, t') = -i\theta(t - t') e^{-i\omega_p^\zeta(t - t')}$. Taking a Markovian approximation for the bath density of states and coupling [i.e assuming that the product of the bath density of states $N^\zeta(\omega)$ and the square of the system–bath coupling $\zeta_{p,0}$ are constant: $\pi \zeta_{p,0}^2 N^\zeta(\omega) = \kappa$] then gives:

$$\sum_p \zeta_{p,0}^2 e^{-i\omega_p^\zeta(t - t')} = 2\kappa \delta(t - t'). \quad (12.19)$$

Putting this all together, the net effect of the decay bath on the self-consistency equation is just to add a decay term, so the result may be written as:

$$(\omega_0 - \mu_S - i\kappa) \psi_0 e^{-i\mu_S t} = - \sum_i \frac{ig_i}{2} \int \frac{d\nu}{2\pi} G_{a_i^\dagger b_i}^K(\nu). \quad (12.20)$$

12.4.2 Pumping bath and $G_{a^\dagger b}^K$

The remaining task is to find the matrix of fermionic Green's functions in the four by four space resulting from the a, b fermionic fields and the $\pm$ space associated with the closed-time-path contour. As above, we will take the interaction Hamiltonian to be the coupling between the system and the bath. This leaves the free fermion Hamiltonian:

$$H_0^{\text{TLS}} = \sum_i \varepsilon_i (b_i^\dagger b_i - a_i^\dagger a_i) + \sum_i g_i \psi_0 \left(e^{i\mu_{st}} a_i^\dagger b_i + e^{-i\mu_{st}} b_i^\dagger a_i \right). \quad (12.21)$$

It is possible to diagonalise this Hamiltonian by a unitary transformation, and thereby write the appropriate free Green's function, however it is first necessary to remove the time dependence introduced by the form of the ansatz for the coherent field. This can be achieved by a gauge transformation:

$$H \rightarrow H - \frac{\mu_S}{2} \left[\sum_i \left(b_i^\dagger b_i - a_i^\dagger a_i \right) + \sum_n \left(B_n^\dagger B_n - A_n^\dagger A_n \right) \right]. \quad (12.22)$$

such that $b \rightarrow b e^{-i\mu_{st}/2}$, $a \rightarrow a e^{i\mu_{st}/2}$, which removes the time dependence of the mean-field photon to fermion coupling. The gauge transformation for the bath modes that also appears in Eq. (12.22) is necessary to ensure no time dependence is introduced into the system-bath coupling terms. The net result is to replace $\varepsilon_i \rightarrow \tilde{\varepsilon}_i = \varepsilon_i - \mu_S/2$ in H_0^{TLS} , and to shift the bath Green's function in frequency by $\pm\mu_S/2$.

After the above transformation, the Hamiltonian can be diagonalised by the unitary transformation:

$$\begin{pmatrix} b_i \\ a_i \end{pmatrix} = \begin{pmatrix} \cos(\theta_i) & \sin(\theta_i) \\ -\sin(\theta_i) & \cos(\theta_i) \end{pmatrix} \begin{pmatrix} \beta_i \\ \alpha_i \end{pmatrix}, \quad (12.23)$$

after which the free Hamiltonian takes the form $H_0^{\text{TLS}} = \sum_i E_i (\beta_i^\dagger \beta_i - \alpha_i^\dagger \alpha_i)$, where $\tan(2\theta_i) = -g_i \psi / \tilde{\varepsilon}_i$ and $E_i^2 = \tilde{\varepsilon}_i^2 + g_i^2 \psi_0^2$. Since the Hamiltonian is diagonal in the β, α basis, the retarded Green's functions in that basis are just $[\nu \mp E_i + i\eta]^{-1}$ (where η is infinitesimal), and so the retarded Green's functions in the b, a basis can be written as:

$$\begin{aligned} G_0^R(\nu) &= \begin{pmatrix} \cos \theta_i & \sin \theta_i \\ -\sin \theta_i & \cos \theta_i \end{pmatrix} \begin{pmatrix} [\nu - E_i + i\eta]^{-1} & 0 \\ 0 & [\nu + E_i + i\eta]^{-1} \end{pmatrix} \begin{pmatrix} \cos \theta_i & -\sin \theta_i \\ \sin \theta_i & \cos \theta_i \end{pmatrix} \\ &= \frac{1}{(\nu + i\eta)^2 - E_i^2} \begin{pmatrix} \nu + \tilde{\varepsilon}_i + i\eta & g_i \psi_0 \\ g_i \psi_0 & \nu - \tilde{\varepsilon}_i + i\eta \end{pmatrix} \end{aligned} \quad (12.24)$$

$$[G_0^R]^{-1} = \begin{pmatrix} \nu - \tilde{\varepsilon}_i + i\eta & -g_i \psi_0 \\ -g_i \psi_0 & \nu + \tilde{\varepsilon}_i + i\eta \end{pmatrix}. \quad (12.25)$$

Just as for the free bosonic Green's functions described in Eq. (12.10), $[G_0^{-1}]^K$ is infinitesimal. Since coupling to the pumping baths, which is discussed next, will add a non-infinitesimal Keldysh self energy, we will neglect this infinitesimal contribution. Therefore, the expression for $[G_0^K]^{-1}$ and its Hermitian conjugate are all that is needed of the free Green's function. What remains is to determine the self energy that arises from coupling to the pumping bath.

Taking the part of the interaction Hamiltonian due to $H_{\text{sys,bath}}^{\text{pump}}$, and inserting it into the definition of U in Eq. (12.8), one has that the interaction vertices are generated by:

$$\begin{aligned} \int_C dt \tilde{H}_{\text{sys,bath}}^{\text{pump}} &= \int_{-\infty}^{\infty} dt \left[\tilde{a}_i^\dagger(t, f) \tilde{A}_n(t, f) - \tilde{a}_i^\dagger(t, b) \tilde{A}_n(t, b) + \dots \right] \\ &= \int_{-\infty}^{\infty} dt \left[\tilde{a}_{i+}^\dagger(t) \tilde{A}_{n-}(t) + \tilde{a}_{i-}^\dagger(t) \tilde{A}_{n+}(t) + \dots \right]. \end{aligned} \quad (12.26)$$

As each pumping bath couples either to only a modes or to only b modes, there are no off diagonal self energy terms in the a, b basis. One may therefore concentrate first on $\Sigma_{a^\dagger a}$, and $\Sigma_{b^\dagger b}$ will follow by analogy. Written as a matrix in the Keldysh space as defined in Eq. (12.7), one has:

$$\Sigma_{a^\dagger a} = \begin{pmatrix} \Sigma_{a^\dagger a}^{++} & \Sigma_{a^\dagger a}^{+-} \\ \Sigma_{a^\dagger a}^{-+} & \Sigma_{a^\dagger a}^{--} \end{pmatrix}. \quad (12.27)$$

The $\pm$ labels determine the label of the incoming/outgoing fields, and it is clear from Eq. (12.26) that $+$ fields couple to $-$ bath fields and vice versa. Thus, an example self energy diagram is: $\Sigma_{a^\dagger a}^{-+} = \text{---} \xrightarrow{a} \text{---} \xrightarrow{A} \text{---} \xrightarrow{a} \text{---}$, giving the equations

$$\begin{aligned} \Sigma_{a^\dagger a}^{++}(t, t') &= \sum_n \Gamma_{i,n}^2 G_{A^\dagger A}^{--} = 0 \\ \Sigma_{a^\dagger a}^{-+}(t, t') &= \sum_n \Gamma_{i,n}^2 G_{A^\dagger A}^{+-} = -i \sum_n \Gamma_{i,n}^2 \theta(t - t') e^{-i\nu_n^\Gamma(t-t')} = -i\gamma\delta(t - t') \\ \Sigma_{a^\dagger a}^{+-}(t, t') &= \sum_n \Gamma_{i,n}^2 G_{A^\dagger A}^{-+} = +i \sum_n \Gamma_{i,n}^2 \theta(t' - t) e^{+i\nu_n^\Gamma(t-t')} = +i\gamma\delta(t - t') \\ \Sigma_{a^\dagger a}^{--}(t, t') &= \sum_n \Gamma_{i,n}^2 G_{A^\dagger A}^{++} = -i \sum_n \Gamma_{i,n}^2 [1 - 2n_A(\nu_n^\Gamma)] e^{-i\nu_n^\Gamma(t-t')} = -2i\gamma\check{F}_A(t - t'). \end{aligned}$$

In the last three lines, the Markovian limit has been taken to give the final equality. In the last line we have used:

$$\check{F}_A(t) = \int \frac{d\nu}{2\pi} e^{-i\nu t} F_A(\nu), \quad F_A(\nu) = 1 - 2n_A(\nu), \quad (12.28)$$

where the form of the distribution function F comes from the form of the equal-time Keldysh Green's function $F_A(\nu_n^\Gamma) = \langle A_n A_n^\dagger - A_n^\dagger A_n \rangle$. As a function of frequency, the

self energy matrix in Keldysh space is thus:

$$\Sigma_{a^\dagger a}(\nu) = \begin{pmatrix} 0 & i\gamma \\ -i\gamma & -2i\gamma F_A(\nu) \end{pmatrix}. \quad (12.29)$$

The matrix for $\Sigma_{b^\dagger b}(\nu)$ is identical except that $F_A(\nu) \rightarrow F_B(\nu)$.

Combining the free Green's function and self energy, we may write the entire inverse Green's function in the basis (b_+, a_+, b_-, a_-) as:

$$G^{-1}(\nu) = \begin{pmatrix} 0 & 0 & \nu - \tilde{\epsilon}_i - i\gamma & -g_i\psi_0 \\ 0 & 0 & -g_i\psi_0 & \nu + \tilde{\epsilon}_i - i\gamma \\ \nu - \tilde{\epsilon}_i + i\gamma & -g_i\psi_0 & 2i\gamma F_B(\nu) & 0 \\ -g_i\psi_0 & \nu + \tilde{\epsilon}_i + i\gamma & 0 & 2i\gamma F_A(\nu) \end{pmatrix}. \quad (12.30)$$

Clearly, if ψ_0 is zero then the a, b fields decouple as expected. However for non-zero ψ_0 , the Keldysh Green's functions of the two fields get mixed, so that their occupation is set by a balance of the pumping and the effects of the coherent photon field.

To complete our analysis, we should invert the above matrix to find the Keldysh block, and the $a^\dagger b$ component of that block. Using the Keldysh block structure of Eq. (12.9), and in particular that $G^K = -G^R[G^{-1}]^K G^A$, one may write:

$$G^K(\nu) = -\frac{2i\gamma}{[(\nu + i\gamma)^2 - E_i^2][(\nu - i\gamma)^2 - E_i^2]} \times \begin{pmatrix} \nu - \tilde{\epsilon}_i + i\gamma & -g_i\psi_0 \\ -g_i\psi_0 & \nu + \tilde{\epsilon}_i + i\gamma \end{pmatrix} \begin{pmatrix} F_B(\nu) & 0 \\ 0 & F_A(\nu) \end{pmatrix} \begin{pmatrix} \nu - \tilde{\epsilon}_i - i\gamma & -g_i\psi_0 \\ -g_i\psi_0 & \nu + \tilde{\epsilon}_i - i\gamma \end{pmatrix}. \quad (12.31)$$

For the mean-field condition in Eq. (12.12), we require in particular the $G_{a_i^\dagger b_i}^K$ component which has the form:

$$G_{a_i^\dagger b_i}^K(\nu) = 2i\gamma g\psi_0 \frac{[F_A(\nu) + F_B(\nu)]\nu + [F_B(\nu) - F_A(\nu)](\tilde{\epsilon}_i + i\gamma)}{[(\nu - E_i)^2 + \gamma^2][(\nu + E_i)^2 + \gamma^2]}. \quad (12.32)$$

12.5 Mean-field theory and its limits

Putting the $G_{a_i^\dagger b_i}^K$ component of Eq. (12.32) into Eq. (12.20) gives the self-consistency condition (equation for the condensate) of the mean-field theory:

$$(\omega_0 - \mu_S - i\kappa)\psi_0 = \sum_i g_i^2 \psi_0 \gamma \int \frac{d\nu}{2\pi} \frac{(F_B + F_A)\nu + (F_B - F_A)(\tilde{\epsilon}_i + i\gamma)}{[(\nu - E_i)^2 + \gamma^2][(\nu + E_i)^2 + \gamma^2]}. \quad (12.33)$$

Equation (12.33) is central to our analysis. This equation is rather powerful, in that it combines several well known theoretical results within a single framework. As

will be shown in section 12.5.1, in the equilibrium limit (where the system–bath couplings are taken to zero) Eq. (12.33) reduces to the gap equation which applies throughout the BCS–BEC crossover. In the opposite highly non-equilibrium limit (see section 12.5.2) it reduces to the standard laser condition. At low densities it reduces to the (complex) Gross-Pitaevskii equation, discussed in section 12.5.4. As such, this approach highlights the connections between these apparently different descriptions of condensates or lasers.

The functions $F_{A,B}$ appearing in Eq. (12.33) were defined as $F_{A,B} = 1 - 2n_{A,B}$, where $n_{A,B}$ are bath occupation functions. These occupations are taken to be externally imposed, and can be chosen to have any form relevant to a particular physical situation. Here, we will choose these to be thermal and at equal temperatures but different chemical potentials. Noting that the fermionic states were supposed to represent two-level systems (or excitons), we take the occupations to satisfy $n_A + n_B = 1$. This therefore requires that we have:

$$F_{A,B}(v) = \tanh \left[\frac{\beta}{2} \left(v \pm \frac{\mu_B - \mu_S}{2} \right) \right], \quad (12.34)$$

where μ_B is an adjustable pumping bath chemical potential, and μ_S appears in this expression due to the shift arising from the gauge transformation in Eq. (12.22). Schematically, this situation is illustrated in Fig. 12.3; one can see that $F_A(-\varepsilon) + F_B(\varepsilon) = 2[1 - n_A(-\varepsilon) - n_B(\varepsilon)] = 0$. Physically, this pumping process is most closely related to electrical pumping. Note that, in the absence of any other processes, contact between the two-level systems (excitons) and the pumping reservoir would control the population of the two-level systems, and so one would have:

$$\langle b^\dagger b - a^\dagger a \rangle = n_B(\varepsilon) - n_A(-\varepsilon) = -\tanh \left[\frac{\beta}{2} \left(\varepsilon - \frac{\mu_B}{2} + \frac{\mu_S}{2} \right) \right].$$

Thus, by pumping with a thermalised bath, one will find a thermalised distribution of excitons. Therefore, in the context of polaritons this pumping scheme resembles closely pumping from a thermalised excitonic reservoir, which is often the case in the experiments.

12.5.1 Equilibrium limit of Mean-field theory

The simplest limit to recover from the non-equilibrium self-consistency equation is that of thermal equilibrium. This corresponds to taking $\gamma, \kappa \rightarrow 0$. Since the self-consistency equation included only the coupling between mean-field photons and the decay bath, there is no way that a thermal distribution can be set by the decay bath. On the other hand, the pumping bath can set a thermal distribution, so to recover a non-trivial equilibrium distribution one should take $\kappa \rightarrow 0$ first, and then $\gamma \rightarrow 0$. If $\kappa = 0$, then the imaginary part of the right hand side of Eq. (12.33) must vanish. In order to satisfy this, without restricting the range of solutions of the real

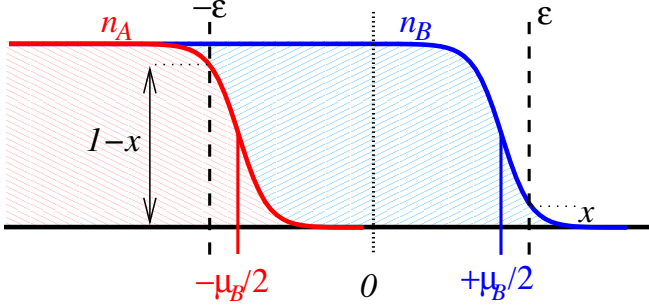


Fig. 12.3 Occupation functions for the pumping baths, chosen to set total occupation of two modes to one, while varying the degree of inversion.

part, one must choose $F_B(v) = F_A(v)$. In terms of the distribution functions written in Eq. (12.34), this clearly means $\mu_S = \mu_B$. Physically, this means that in the absence of decay, the chemical potential of the condensate matches the pumping bath.

After fixing μ_S , the remaining part of the equation becomes:

$$(\omega_0 - \mu_B)\psi_0 = \sum_i g_i^2 \psi_0 \gamma \int \frac{dv}{2\pi} \frac{2 \tanh(\beta v/2) v}{[(v - E_i)^2 + \gamma^2][(v + E_i)^2 + \gamma^2]}. \quad (12.35)$$

We may then take the limit of small γ , by using:

$$\lim_{\gamma \rightarrow 0} \frac{2\gamma v}{[(v - E_i)^2 + \gamma^2][(v + E_i)^2 + \gamma^2]} = \frac{2\pi}{4E_i} [\delta(v - E_i) - \delta(v + E_i)], \quad (12.36)$$

hence we find:

$$\begin{aligned} (\omega_0 - \mu_B)\psi_0 &= \sum_i \frac{g_i^2 \psi_0}{4E_i} \int dv \tanh\left(\frac{\beta v}{2}\right) [\delta(v - E_i) - \delta(v + E_i)] \\ &= \sum_i \frac{g_i^2 \psi_0}{2E_i} \tanh\left(\frac{\beta E_i}{2}\right). \end{aligned} \quad (12.37)$$

This is the equilibrium result[51, 52, 53], but with the two-level constraint on the fermions imposed only on average.¹ Note, that this is the standard mean-field gap equation of the BCS-BEC crossover theory [56].

¹ Imposing the two-level constraint on average, the equilibrium expectation of the inversion $\langle b^\dagger b - a^\dagger a \rangle$ can be written as:

$$\frac{e^{\beta E} - e^{-\beta E}}{1 + e^{\beta E} + e^{-\beta E} + 1} = \frac{(e^{\beta E/2} - e^{-\beta E/2})(e^{\beta E/2} + e^{-\beta E/2})}{(e^{\beta E/2} + e^{-\beta E/2})^2} = \tanh\left(\frac{\beta E}{2}\right). \quad (12.38)$$

Were the two-level constraint imposed exactly, the result would instead be: $(e^{\beta E} - e^{-\beta E})/(e^{\beta E} + e^{-\beta E}) = \tanh(\beta E)$, as the zero and doubly occupied states would be removed from the denominator.

12.5.2 High temperature limit of Mean-field theory - simple laser

The opposite extreme to the equilibrium condensate is the limit of a simple laser, which can also be recovered from Eq. (12.33). Before showing how this limit can be recovered from our theory, we first provide a brief summary of the threshold condition of a simple laser, and express it in similar language to the above self-consistency condition. The equations describing the steady state of a laser can be derived starting from the well-known Maxwell-Bloch equations:

$$\partial_t \psi_0 = -i\omega_0 \psi_0 - \kappa \psi_0 + \sum_i g_i P_i, \quad (12.39)$$

$$\partial_t P_i = -2i\varepsilon_i P - \lambda_\perp P_i + g_i \psi_0 N_i \quad (12.40)$$

$$\partial_t N_i = \lambda_\parallel (N_0 - N_i) - 2g_i (\psi_0^* P_i + P_i^* \psi_0). \quad (12.41)$$

These equations can be understood as originating from considering a Hamiltonian like Eq. (12.1), with $P_i = -i\langle a_i^\dagger b_i \rangle$, $N_i = \langle b_i^\dagger b_i - a_i^\dagger a_i \rangle$. One then writes the Heisenberg-Langevin equations, with a Markovian set of baths distinct for each two-level system, and then takes the semiclassical approximation to drop bath noise operators. The value N_0 is the bath inversion imposed by the pumping. Note that with coupling to such a Markovian pumping bath, there is a discontinuous jump between the allowed steady states with no decay, and the laser-like solutions found for any non-zero pumping[57]. In particular, with pumping and decay, inversion is always required for a condensed solution of these Maxwell-Bloch equations, so they cannot smoothly interpolate between a condensate and a laser. Such behaviour should not be too surprising, as a frequency independent (Markovian) bath occupation corresponds to an infinite temperature, and so even arbitrarily weak coupling of the system to an infinite temperature reservoir may destroy the condensate. With more realistic models of pumping, such a discontinuous jump need not necessarily occur. One should thus interpret the microscopic origin and consequent behaviour of Eqs. (12.39 - 12.41) with some caution. However, since Maxwell-Bloch equations of the above form are frequently used as a simple model of a laser, it is instructive to see what approximations they would correspond to in terms of our non-equilibrium formalism, in which the microscopic description of the pumping is better controlled.

Starting from these Maxwell-Bloch equations, the self-consistency condition for a macroscopic photon field $\psi_0(t) = \psi_0 e^{-i\mu t}$ can be written as:

$$(-i\mu + i\omega_0 + \kappa) \psi_0 = \sum_i g_i P_i, \quad (-i\mu + 2i\varepsilon_i + \lambda_\perp) P_i = g_i \psi_0 N_i, \quad (12.42)$$

which can be combined to write a single self-consistency condition:

$$(\omega_0 - \mu - i\kappa) \psi_0 = - \sum_i \frac{g_i^2 \psi_0 N_i}{2\varepsilon_i - i\lambda_\perp}. \quad (12.43)$$

Substituting the steady state value of P_i from Eq. (12.42) into Eq. (12.41) gives:

$$N_0 = N_i \left[1 + \frac{2g_i^2 |\psi_0|^2}{\lambda_{\parallel}} \frac{2\lambda_{\perp}}{\lambda_{\perp}^2 + 4\tilde{\epsilon}_i^2} \right] \quad (12.44)$$

hence we may substitute this into Eq. (12.43) to give the final form of the self-consistency condition for the Maxwell-Bloch equations:

$$(\omega_0 - \mu - i\kappa) \psi_0 = - \sum_i g_i^2 \psi_0 N_0 \frac{(2\tilde{\epsilon}_i + i\lambda_{\perp})}{[4\tilde{\epsilon}_i^2 + \lambda_{\perp}^2 + 4(\lambda_{\perp}/\lambda_{\parallel})g_i^2 |\psi_0|^2]}. \quad (12.45)$$

The laser threshold condition is given by taking $\psi \rightarrow 0$ in the above equation. If we also take $g_i = g$, $\epsilon_i = \epsilon$, and the usual laser operating condition of $\lambda_{\perp} \gg \kappa$ one has that lasing occurs at the cavity frequency, $\mu = \omega_0$ and the threshold condition has the well-known form: $\kappa \lambda_{\perp} / g^2 = n N_0$, where n is the number of two-level systems.

12.5.2.1 Recovering laser limit from non-equilibrium mean-field theory

This simple laser self-consistency condition can be recovered from equation (12.33) if rather than using the frequency dependent forms for $F_{A,B}(\nu)$ discussed previously, one instead takes $F_{A,B}$ to be constants. Physically such a limit corresponds to high temperatures. Note that as the temperature rises, to keep the bath population fixed, the chemical potential must also vary. We will therefore take $\mu \propto T$, and then take the limit $T \rightarrow \infty$. Such a limit has another simple interpretation, corresponding to making a fully Markovian approximation, including assuming the occupation, as well as the density of states, to be flat, and so writing the Keldysh part of the self energy as

$$\Sigma_{a^{\dagger}a}^{-}(t, t') = -i \sum_n \Gamma_{i,n}^2 [1 - 2n_F(\nu_n^{\Gamma})] e^{-i\nu_n^{\Gamma}(t-t')} = -2i\gamma F_A \delta(t-t'). \quad (12.46)$$

As such, our approach in Eq. (12.29) is Markovian for the density of states of the bath, but non-Markovian for the occupation. In terms of quantum statistical (i.e. Heisenberg-Langevin) approaches, the distinction is whether the noise should be taken as white noise or coloured noise. Assuming the noise correlations to be white, and thus neglecting the frequency dependence of occupation, is also the approximation underlying the quantum regression theorem[49], which allows one to relate two-time correlations to the evolution of the density matrix. The role of this approximation, and its implications for the fluctuation dissipation theorem are discussed by Ford and O'Connell[50].

If $F_{A,B}$ are frequency independent, then in Eq. (12.33), the term in the integral proportional to ν will vanish as this is an odd function, and so Eq. (12.33) becomes:

$$(\omega_0 - \mu - i\kappa) \psi_0 = \sum_i g_i^2 \psi_0 (F_B - F_A) \frac{\tilde{\epsilon}_i + i\gamma}{4(E_i^2 + \gamma^2)}. \quad (12.47)$$

Hence, the polarisation of the two-level systems is in this case proportional to the inversion of the baths, $N_0 = (n_B - n_A) = -(F_B - F_A)/2$ and we have:

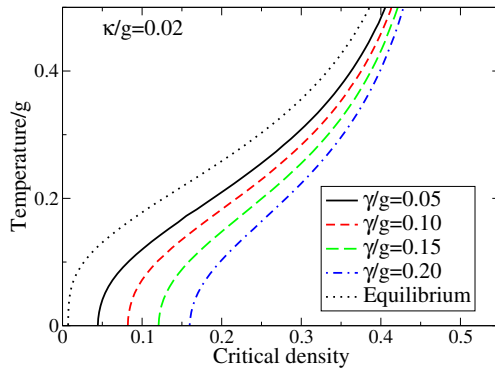
$$(\omega_0 - \mu - i\kappa)\psi_0 = -\sum_i g_i^2 \psi_0 N_0 \frac{\tilde{\epsilon}_i + i\gamma}{2(E_i^2 + \gamma^2)}. \quad (12.48)$$

Then, identifying the decay constants in Eq. (12.45) as $\lambda_\perp = \lambda_\parallel = 2\gamma$, Eq. (12.48) and Eq. (12.45) are equivalent.

12.5.3 General properties of mean-field theory away from extremes

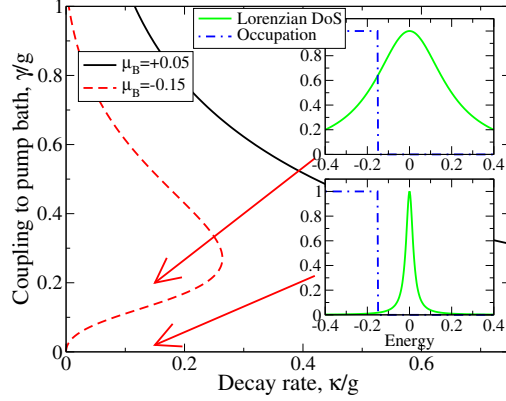
Away from the extremes of laser theory or of thermal equilibrium, the effect of pumping on the phase boundary can be understood as a result of competition of two effects: pumping and decay add noise, reducing coherence, hence suppressing condensation; on the other hand, for a given decay rate, pumping increases the density, favouring condensation. The simplest illustration of the first of these is shown in Fig. 12.4, where one sees that as the value of γ is increased, for a fixed κ , the critical density required for condensation increases.

Fig. 12.4 Critical temperature as a function of density, showing effects of pumping and decay, taking a Gaussian distribution of two-level-system energies with variance 0.15g. Adapted from Ref.[29].



To see the competition between pumping causing dephasing and pumping increasing density, one may look at the low temperature limit, shown in Fig. 12.5, plotting the critical value of κ as a function of γ . Two lines are shown; the solid line has an inverted bath (as would be required for the laser limit), the dashed line has a non-inverted bath. In the later case (as illustrated in the inset) for small γ , the two-level system energy is too far below the pumping bath, and insufficiently broadened by γ , to be populated; for larger γ the broadening is sufficient, and condensation may occur. In the presence of inhomogeneous broadening, the above picture is significantly relaxed, since the tail of the density of states can be occupied even if the peak is below the chemical potential.

Fig. 12.5 Critical couplings to pumping bath without inhomogeneous broadening and at low temperatures.



12.5.4 Low density limit: recovering complex Gross-Pitaevskii equation

The self-consistency condition of Eq. (12.33) can also be related to the idea of the complex Gross-Pitaevskii equation providing a mean-field description of a spatially varying condensate. For a steady uniform state, the mean-field self-consistency condition may be understood as $(\mu_S + i\kappa - \omega_0)\psi_0 = \chi[\psi_0, \mu_S]\psi_0$, where $\chi[\psi_0, \mu_S]$ is a nonlinear complex susceptibility. For a $\psi_0(r, t)$ which varies slowly in space and time [up to an allowed fast time dependence described by a factor $\exp(-i\mu_S t)$], one may consider the local density approximation:

$$\left(i\partial_t + i\kappa - \left[V(r) - \frac{\nabla^2}{2m} \right] \right) \psi_0(r, t) = \chi[\psi_0(r, t)]\psi_0(r, t). \quad (12.49)$$

In order to determine the large scale spatial structure of a condensate, or its low energy collective modes, it is often sufficient to make a Taylor expansion of the nonlinear complex susceptibility, resulting in a complex Gross-Pitaevskii equation:

$$i\partial_t \psi_0 = \left(-\frac{\nabla^2}{2m} + V(r) + U|\psi_0|^2 + i[\gamma_{\text{eff}}(\mu_B) - \kappa - \Gamma|\psi_0|^2] \right) \psi_0, \quad (12.50)$$

where Γ represents the simplest form of nonlinearity of the imaginary part, taking a form that will ensure stability.

Depending on the details of pumping included in the model, one may find that by treating $\chi[\psi(t)]$ more carefully the susceptibility depends not only on the current value of $\psi(t)$, but on its history, due to dynamics of the reservoir. [In fact, to correctly reproduce the polariton spectrum, one ought to take the excitonic susceptibility to have a resonance at the exciton energy, after which a variant of Eq. (12.50), but with the appropriate polariton dispersion will be recovered.] In the limit of sufficiently slow dynamics of the system, or when considering steady

states, dynamics of the reservoir should become unimportant. Results of the complex Gross-Pitaevskii equation with or without separate reservoir dynamics may be found elsewhere [45, 46, 47] and are discussed in section 11.5 of Chapter 11.

12.6 Fluctuations, and instability of the normal state

As stated earlier, when introducing the self-consistency condition for the non-equilibrium problem, it is not possible to consider minimising free energy when looking at a system coupled to multiple baths, and so it is instead necessary to look for stable steady states. The self consistency conditions discussed above determine whether a steady state may exist, but not whether it is stable. In order to analyse stability, it is necessary to consider fluctuations about a given state, and to find whether they grow or decay in time. In addition, the study of fluctuations allows one to determine the response functions of the system — in the present context, this means the photon Green's function — which in turn will give the physical observables, such as photoluminescence and absorption spectra.

In the non-equilibrium case, both the spectrum of possible excitations (i.e. what is seen in the absorption spectrum), and its occupation (i.e. photoluminescence) must be determined independently, for which the Keldysh Green's function approach is ideal. In the following, the approach to calculating these Green's functions is discussed for both the normal and condensed state, and then this approach is applied to understanding the instability of the normal state, which allows a clearer interpretation of the relation between the non-equilibrium condensate and a simple laser. For the condensed system, the calculations are more complicated due to the existence of non-zero anomalous correlations, i.e. $\langle \psi_k(t) \psi_{-k}(t') \rangle$; the general structure of the spectrum of the non-equilibrium system will be discussed in section 12.7.

12.6.1 Photon Green's functions in the non-equilibrium model

To allow for anomalous correlations in the condensed state, it is helpful to write the Green's function in a vector space of $\psi_k, \psi_{-k}^\dagger$. Just as in the above discussion of the Green's functions for the two-level system, this vector space of $\psi_k, \psi_{-k}^\dagger$ should be combined with the $\pm$ space due to the Keldysh/retarded/advanced structure. Thus, one has four by four matrices, in the basis $(\psi_{k,+}, \psi_{-k,+}^\dagger, \psi_{k,-}, \psi_{-k,-}^\dagger)$.

The photon Green's function can be found by solving the Dyson equation, $D^{-1} = D_0^{-1} - \Sigma$, and so to start with, the free photon Green's function is required. The free Hamiltonian in this case is just $H_0^{\text{photon}} = \sum_k \omega_k \psi_k^\dagger \psi_k$. In the four by four basis arising from mixing $\psi_k, \psi_{-k}^\dagger$, some elements correspond to Green's functions in which $\psi, \psi^\dagger$ are interchanged in order. This means that these elements are Hermitian conjugated, giving the form:

$$D_0^{-1} = \begin{pmatrix} 0 & 0 & \omega - \tilde{\omega}_k - i\eta & 0 \\ 0 & 0 & 0 & -\omega - \tilde{\omega}_k + i\eta \\ \omega - \tilde{\omega}_k + i\eta & 0 & (2i\eta)F_0(\omega + \mu) & 0 \\ 0 & -\omega - \tilde{\omega}_k - i\eta & 0 & (2i\eta)F_0(-\omega + \mu) \end{pmatrix}, \quad (12.51)$$

where once again η is infinitesimal. In this, we have written all frequencies measured relative to μ_S , meaning that we made the substitution $\psi_k \rightarrow e^{-i\mu_S t}(\psi_0\delta_{k,0} + \psi_k)$.

To this free Green's function one must add self energies arising from two parts of the interaction Hamiltonian. The first is the coupling between cavity photons and the decay bath; the second is the coupling between the photons and the pumped two-level systems. The first contribution has a form exactly analogous to the coupling between the two-level systems and pumping baths, i.e.:

$$\begin{aligned} \Sigma_{\psi^\dagger\psi}^{++}(t, t') &= \sum_p \zeta_{p,k}^2 D_{\psi^\dagger\psi}^{--} = 0 \\ \Sigma_{\psi^\dagger\psi}^{+-}(t, t') &= \sum_p \zeta_{p,k}^2 D_{\psi^\dagger\psi}^{+-} = -i \sum_p \zeta_{p,k}^2 \theta(t-t') e^{-i\omega_p^\zeta(t-t')} = -i\kappa\delta(t-t') \\ \Sigma_{\psi^\dagger\psi}^{-+}(t, t') &= \sum_p \zeta_{p,k}^2 D_{\psi^\dagger\psi}^{-+} = +i \sum_p \zeta_{p,k}^2 \theta(t'-t) e^{+i\omega_p^\zeta(t-t')} = +i\kappa\delta(t-t') \\ \Sigma_{\psi^\dagger\psi}^{--}(t, t') &= \sum_p \zeta_{p,k}^2 D_{\psi^\dagger\psi}^{++} = -i \sum_p \zeta_{p,k}^2 [2n_\psi(\omega_p^\zeta) + 1] e^{-i\omega_p^\zeta(t-t')} = -2i\kappa\check{F}_\psi(t-t'), \end{aligned}$$

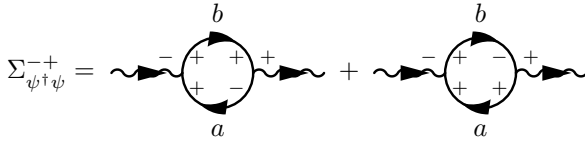
where as before $\check{F}_\psi$ is the Fourier transform of the $2n_\psi(\omega) + 1$, and the Markovian limit for the bath density of states and coupling constant has been applied to get the final expression; these terms thus give a self energy:

$$\Sigma_{\text{decay}}(\omega) = \begin{pmatrix} 0 & 0 & +i\kappa & 0 \\ 0 & 0 & 0 & -i\kappa \\ -i\kappa & 0 & -(2i\kappa)F_\psi(\omega + \mu_S) & 0 \\ 0 & +i\kappa & 0 & -(2i\kappa)F_\psi(-\omega + \mu_S) \end{pmatrix}. \quad (12.52)$$

In calculating the self energy due to the coupling to two-level systems, one may simplify the calculation by noting that only $\Sigma_{\psi^\dagger\psi}^R, \Sigma_{\psi^\dagger\psi}^R, \Sigma_{\psi^\dagger\psi}^K, \Sigma_{\psi^\dagger\psi}^K$ are independent; all other self energies can be related to these quantities by Hermitian conjugation and/or swapping $\omega \rightarrow -\omega$. To generate the diagrams for these self energies, we should first determine the interaction vertices that give rise to such self energies. The relevant part of the interaction Hamiltonian here is the interaction between two-level systems and incoherent photons, and so the relevant contribution to U comes from

$$\begin{aligned}
\int_C dt \tilde{H}_{\text{int}}^{\text{TLS-photon}} &= \int_{-\infty}^{\infty} dt g \left[\tilde{\psi}(t, f) \tilde{b}_i^\dagger(t, f) \tilde{a}_i(t, f) - \tilde{\psi}(t, b) \tilde{b}_i^\dagger(t, b) \tilde{a}_i(t, b) + \text{H.c.} \right] \\
&= \int_{-\infty}^{\infty} dt \frac{g}{\sqrt{2}} \left[\tilde{\psi}_+(t) \left(\tilde{b}_{i+}^\dagger(t) \tilde{a}_{i-}(t) + \tilde{b}_{i-}^\dagger(t) \tilde{a}_{i+}(t) \right) \right. \\
&\quad \left. + \tilde{\psi}_-(t) \left(\tilde{b}_{i+}^\dagger(t) \tilde{a}_{i+}(t) + \tilde{b}_{i-}^\dagger(t) \tilde{a}_{i-}(t) \right) + \text{H.c.} \right].
\end{aligned} \tag{12.53}$$

The self energy diagrams thus consist of diagrams with one incoming and one outgoing photon line, connected via the interaction vertices in Eq. (12.53), and the Green's functions for the two-level system. As is clear from Eq. (12.53), the vertices all involve the two-level system swapping between the a and b states. Just as for the diagrams describing the effects of the bath discussed in Sec. 12.4, one must also keep track of the $\pm$ labels on the fields. To calculate, for instance, the retarded self energy (i.e. the $-+$ component) it is clear that the vertices arising from the possible placements of $\pm$ signs have the form:



(any other set of possible $\pm$ labels on the internal lines will involve a $--$ line, and such Green's functions vanish). To translate these diagrams into an equation for the self energy, one must use the following Feynman rules (see Refs.[31, 33, 32]): For each interaction vertex there is a factor $(-ig/\sqrt{2})$, and for each internal Green's function, a factor iG . There is then a prefactor $i(-1)^F$, where F is the number of closed Fermion loops ($F = 1$ in the current case), and there is a combinatoric factor associated with how the vertices are found from the expansion of U , which is the same as in any other diagrammatic approach. Applying these rules, one may write:

$$\Sigma_{\psi^\dagger\psi}^{-+} = -i \frac{2}{2!} \left(\frac{g}{\sqrt{2}} \right)^2 \int \frac{d\nu}{2\pi} \sum_i \left[G_{a_i^\dagger a_i}^A(\nu) G_{b_i^\dagger b_i}^K(\nu + \omega) + G_{a_i^\dagger a_i}^K(\nu) G_{b_i^\dagger b_i}^R(\nu + \omega) \right]. \tag{12.54}$$

For the anomalous case, all that changes is the a, b labels, i.e.:

$$\Sigma_{\psi^\dagger\psi^\dagger}^{-+} = -i \frac{2}{2!} \left(\frac{g}{\sqrt{2}} \right)^2 \int \frac{d\nu}{2\pi} \sum_i \left[G_{a_i^\dagger b_i}^A(\nu) G_{b_i^\dagger a_i}^K(\nu + \omega) + G_{a_i^\dagger b_i}^K(\nu) G_{b_i^\dagger a_i}^R(\nu + \omega) \right]. \tag{12.55}$$

The component Σ^{+-} is just the Hermitian conjugate of Σ^{-+} as above. The component Σ^{++} vanishes, since it either involves $--$ lines, or it involves products of two retarded Green's functions. Since the retarded Green's function is causal — i.e. $D^R(t, t') \propto \theta(t - t')$ — then as a function of frequency, all of its poles are in the lower half plane, and so the integral of a product of two such functions is equal to

zero.² The only other surviving component of the self energy is thus:

$$\Sigma_{\psi^\dagger\psi}^{--} = \text{diagram 1} + \text{diagram 2} + \text{diagram 3}$$

which gives the equation:

$$\Sigma_{\psi^\dagger\psi}^{--} = -i \frac{2}{2!} \left(\frac{g}{\sqrt{2}} \right)^2 \int \frac{d\nu}{2\pi} \sum_i \left[G_{a_i^\dagger a_i}^K(\nu) G_{b_i^\dagger b_i}^K(\nu + \omega) + G_{a_i^\dagger a_i}^A(\nu) G_{b_i^\dagger b_i}^R(\nu + \omega) \right. \\ \left. + G_{a_i^\dagger a_i}^R(\nu) G_{b_i^\dagger b_i}^A(\nu + \omega) \right]. \quad (12.56)$$

As was the case for the retarded components, the only difference between normal and anomalous Keldysh components is in the a, b labels, so:

$$\Sigma_{\psi^\dagger\psi^\dagger}^{--} = -i \frac{2}{2!} \left(\frac{g}{\sqrt{2}} \right)^2 \int \frac{d\nu}{2\pi} \sum_i \left[G_{a_i^\dagger b_i}^K(\nu) G_{b_i^\dagger a_i}^K(\nu + \omega) + G_{a_i^\dagger b_i}^A(\nu) G_{b_i^\dagger a_i}^R(\nu + \omega) \right. \\ \left. + G_{a_i^\dagger b_i}^R(\nu) G_{b_i^\dagger a_i}^A(\nu + \omega) \right]. \quad (12.57)$$

Combining the self energies due to the pumped two-level systems and the self energy due to decay with the free inverse Green's function, one can then find expressions for the photon Green's functions, and hence observable quantities such as the photoluminescence intensity as a function of frequency and momentum, which is given by $\mathcal{L}(\omega) = i \left(D_{\psi^\dagger\psi}^K - D_{\psi^\dagger\psi}^R + D_{\psi^\dagger\psi}^A \right) / 2$. In section 12.6.2, the normal state Green's functions are studied: we show how an effective density of states and occupation function can be defined, and also show how the behaviour of these functions can be related to the structure of the inverse Green's function, and to the stability of the normal system.

In the condensed state, just as in equilibrium, the form of the inverse Green's function can be shown to obey the Hugenholtz-Pines relation[58] (see also [59, Chapter 6]), meaning that $[D_{\psi^\dagger\psi}^R]^{-1}(0,0) = [D_{\psi^\dagger\psi^\dagger}^R]^{-1}(0,0)$, which implies there is a gapless spectrum. Just as in equilibrium, one may show that the requirement for the Hugenholtz-Pines relation to be satisfied is equivalent to the mean-field condition, Eq. (12.33). It is worth noting that as $\psi_0 \rightarrow 0$, the Hugenholtz-Pines relation (and hence the mean-field condition) become equivalent to the condition that:

$$[D_{\psi^\dagger\psi}^R]^{-1}(\omega = \mu_{\text{eff}}, k = 0) = \mu_{\text{eff}} - \omega_0 + i\kappa - \Sigma_{\text{TLS}}^R(\mu_{\text{eff}}) = 0, \quad (12.58)$$

² NB; since the Green's function generically looks like $1/\omega$ at large ω , the integral of a single retarded Green's function depends on the regularisation used. However, for a product of retarded Green's functions, the integral is well defined, and so vanishes.

for some particular μ_{eff} . (In this expression, the non-condensed self energies have been written without the gauge transform of Eq. (12.22), as in the absence of a condensate, there is no reason to perform the gauge transformation.) Section 12.6.2 will show that the condition in Eq. (12.58) also corresponds to the point when the normal state ceases to be stable.

12.6.2 Normal-state Green's functions and instability

Focusing on the non-condensed case, the properties of the spectrum are entirely determined by three real functions of ω , as one may write:

$$\left[D_{\psi^\dagger\psi}^R \right]^{-1}(\omega) = A(\omega) + iB(\omega), \quad \left[D_{\psi^\dagger\psi}^{-1} \right]^K(\omega) = iC(\omega). \quad (12.59)$$

The forms of $A(\omega), B(\omega), C(\omega)$ follow from the expressions in the previous section. These somewhat simplify since we are considering the normal case, and so one has:

$$\left[D_{\psi^\dagger\psi}^R \right]^{-1}(\omega) = \omega - \omega_k + i\kappa - \Sigma_{\text{TLS}}^R(\omega), \quad \left[D_{\psi^\dagger\psi}^{-1} \right]^K(\omega) = 2i\kappa F_\psi(\omega) - \Sigma_{\text{TLS}}^K(\omega), \quad (12.60)$$

where $\Sigma_{\text{TLS}}^{R,K}$ are the self energies from the pumped two-level systems given by Eq. (12.54) and Eq. (12.56). In the following we will first discuss how the forms of $A(\omega), B(\omega), C(\omega)$ determine the spectrum, occupation and stability, and then illustrate this with their forms arising from the particular microscopic model discussed above.

Inverting the matrix of Keldysh Green's functions (using Eq. (12.9)), one finds:

$$D_{\psi^\dagger\psi}^R(\omega) = \frac{A(\omega) - iB(\omega)}{A(\omega)^2 + B(\omega)^2}, \quad D_{\psi^\dagger\psi}^K(\omega) = \frac{-iC(\omega)}{A(\omega)^2 + B(\omega)^2}, \quad (12.61)$$

and then in terms of these quantities, we may write the luminescence spectrum:

$$\mathcal{L}(\omega) = \frac{i}{2} \left[D_{\psi^\dagger\psi}^K(\omega) - \left(D_{\psi^\dagger\psi}^R(\omega) - D_{\psi^\dagger\psi}^A(\omega) \right) \right] = \frac{C(\omega) - 2B(\omega)}{2[A(\omega)^2 + B(\omega)^2]}. \quad (12.62)$$

Further, by analogy with the equilibrium system, we can explain the form of this expression in terms of a spectral weight (density of states) $\rho(\omega) = -2\Im[D_{\psi^\dagger\psi}^R(\omega)]$ and an occupation function $2n_\psi(\omega) + 1 = iD_{\psi^\dagger\psi}^K(\omega)/\rho(\omega)$, giving:

$$\rho(\omega) = \frac{2B(\omega)}{A(\omega)^2 + B(\omega)^2}, \quad n_\psi(\omega) = \frac{1}{2} \left[\frac{C(\omega)}{2B(\omega)} - 1 \right], \quad (12.63)$$

hence the luminescence is related to these as $\mathcal{L}(\omega) = \rho(\omega)n_\psi(\omega)$ as expected.

In the absence of coupling to the two-level systems (and hence neglecting $\Sigma_{\text{TLS}}^{R,K}$ in Eq. (12.60), one may clearly identify the role of the three expressions involved here:

- $B(\omega) = \kappa$ is the linewidth of the normal modes
- $A(\omega) = \omega - \omega_k$ describes the locations of these modes, and
- $C(\omega) = 2\kappa(2n_\psi + 1)$ describes their occupation.

However, when coupled to the two-level systems, $B(\omega)$ is not a constant, hence firstly, the linewidth varies, and more importantly, $B(\omega)$ may vanish at some value of ω . If $B(\omega)$ does vanish then the occupation diverges, but since the spectral weight vanishes too, the luminescence remains finite.

Physically, this describes the behaviour that would, in equilibrium, be expected at the chemical potential, as long as the chemical potential lies below the bottom of the band. Note that the equilibrium Bose-Einstein distribution diverges at the chemical potential. However, if the chemical potential lies below the bottom of the band then the spectral weight is zero at the chemical potential and thus the particle number (luminescence) remains finite.

Out of equilibrium, the system distribution may in general be far from the Bose-Einstein distribution. Even so, when near the threshold for condensation, the system distribution shares an important property with the Bose-Einstein distribution: near the frequency where the condensate will emerge [i.e near the point where $B(\omega) = 0$] the system distribution will diverge as $1/(\omega - \mu_{\text{eff}})$, just as the Bose-Einstein distribution does. We may thus identify the effect of pumping as introducing a chemical potential that has nothing to do with the chemical potential of the decay bath. Since $B(\omega)$ is given by the inverse retarded Green's function, one may note that the inverse Keldysh Green's function does not on its own fix the distribution; it is the ratio of Keldysh and imaginary retarded Green's functions that matter. Figure 12.6 shows how the spectral weight, occupation and luminescence are related to the zeros of the real and imaginary parts of the inverse Green's function.

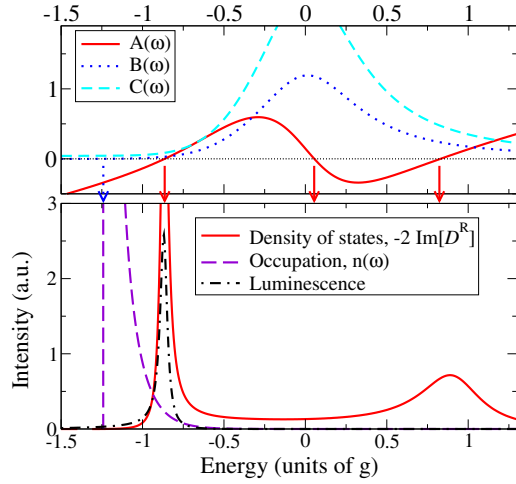
12.6.2.1 Zeros of $A(\omega), B(\omega)$ and stability

Although a zero of $B(\omega)$ alone does not cause the luminescence to diverge, a simultaneous zero of $A(\omega)$ and $B(\omega)$ will. The stability of the system can be seen to change when this occurs, as will be discussed next. When near a simultaneous zero, one may expand $A(\omega) = \alpha(\omega - \xi)$, and $B(\omega) = \beta(\omega - \mu_{\text{eff}})$, and so:

$$\begin{aligned} [D_{\psi^\dagger\psi}^R]^{-1}(\omega) &\simeq \alpha(\omega - \xi) + i\beta(\omega - \mu_{\text{eff}}) \\ &= (\alpha + i\beta) \left[\omega - \frac{(\alpha\xi + i\beta\mu_{\text{eff}})(\alpha - i\beta)}{\alpha^2 + \beta^2} \right], \end{aligned} \quad (12.64)$$

hence the actual poles are at frequencies:

Fig. 12.6 Behaviour of inverse Green's functions, and resulting properties of spectral weight, luminescence and occupation functions in the normal state. Upper panel shows the inverse Green's functions (with zeros marked by arrows), and the lower panel shows the various physical correlations of interest. Adapted from Ref.[29].



$$\omega^* = \frac{(\alpha^2 \xi + \beta^2 \mu_{\text{eff}}) + i\alpha\beta(\mu_{\text{eff}} - \xi)}{\alpha^2 + \beta^2}. \quad (12.65)$$

These poles determine the time dependence of the retarded Green's function, so if $\mu_{\text{eff}} > \xi$, then the pole has the wrong sign of imaginary part and the normal state is unstable. When $\mu_{\text{eff}} = \xi$, then this means there is a value $\omega = \mu_{\text{eff}} = \xi$ for which $[D_{\psi^\dagger \psi}^R]^{-1}(\omega, k=0) = 0$, which as discussed in Eq. (12.58) is equivalent to saying that the mean-field consistency condition can be satisfied. Hence, instability of the normal state, and the existence of a condensed solution will occur together.

It is helpful here to explicitly write $B(\omega)$, in order to understand the origin of its zeros, and what parameters determine their location. In the non-condensed case, the fermionic Green's functions that come from inverting Eq. (12.30) have a simple form:

$$G_{b^\dagger b, a^\dagger a}^R = \frac{1}{v \mp \varepsilon_i + i\gamma}, \quad G_{b^\dagger b, a^\dagger a}^K = -\frac{2i\gamma F_{B,A}(v)}{(v \mp \varepsilon_i)^2 + \gamma^2}, \quad (12.66)$$

and so substituting these into Eq. (12.54), and taking the imaginary part one may write:

$$B(\omega) = \kappa + \gamma^2 \int \frac{dv}{2\pi} \sum_i g_i^2 \frac{F_B(v + \omega) - F_A(v)}{[(v + \omega - \varepsilon_i)^2 + \gamma^2] [(v + \varepsilon_i)^2 + \gamma^2]}. \quad (12.67)$$

For $B(\omega)$ to have zeros, it is necessary that the second term (which originates from pumping) should be negative, and should overcome the first term (which originates from decay). With $F_{A,B}(v) = \tanh[\beta(v \pm \mu_B/2)/2]$, it is clear that this criterion requires μ_B to be sufficiently large. As such, the following scenario describes what happens as μ_B is increased:

Very weak pumping. For large negative μ_B , one finds that $F_B(v + \omega) - F_A(v)$ is always positive, and so no zero of $B(\omega)$ exists.

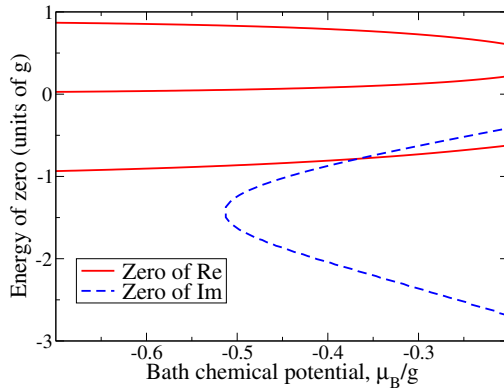
Subcritical pumping. For less negative values of μ_B , there is a range of ω for which $B(\omega)$ is negative, indicating a range of gain in the spectrum. The boundary of this region, where $B(\omega) = 0$ defines an effective chemical potential μ_{eff} , but since $\mu_{\text{eff}} < \xi$ the normal state remains stable.

Critical pumping. At some value of μ_B , one finds that $\mu_{\text{eff}} = \xi$, meaning that at this value of $\omega^* = \mu_{\text{eff}} = \xi$, one has $D_{\psi^\dagger \psi}^R(\omega^*) = 0$. Hence, the gap equation first has a solution at this point, there is a real divergence of the luminescence, and the normal state is marginally stable.

Supercritical pumping. Above this critical value of μ_B , the normal state would have $\mu_{\text{eff}} > \xi$, and so would be unstable.

The actual behaviour for the polariton model of Eq. (12.1) is shown in Fig. 12.7; one can see that a pair of zeros of the imaginary part emerge, and then one crosses the bottom of the polariton modes. Note that in equilibrium, we have $\mu_{\text{eff}} = \mu = \mu_B$ at all conditions, and so only the last three stages of the above scenario exist; condensation occurs when the chemical potential reaches the bottom of the band. It is also important to note that the above scenario means that the Bose-Einstein distribution is not the only distribution that would allow condensation. Any distribution which has the above property, i.e. a divergence at some frequency for given values of the control parameters (density, coupling constant, etc.), is sufficient for quantum condensation in bosonic systems.

Fig. 12.7 Variation of energies of zeros of real part of inverse Green's function and imaginary part as density is varied via chemical potential of pumping bath. The three solid lines correspond to (from the bottom) lower polariton, exciton, and upper polariton respectively. The point where the dashed line crosses solid line is where the condensation occurs. Adapted from Ref.[29].



12.6.2.2 Simplified form of distribution function in high-temperature limit

The way in which the effective distribution is set by the balance of pumping and decay can be demonstrated more clearly by specialising to the case of $\gamma \ll T$, for

which the pumping bath occupation functions do not change significantly across each Lorentzian broadened peak (but may vary between the two peaks). In addition, consider taking $g_i = g, \varepsilon_i = \varepsilon$, so that sums of two-level systems can be replaced by factors n . Then the expression in Eq. (12.67) can be simplified to give:

$$B(\omega) = \kappa + ng^2\gamma \frac{[F_B(\varepsilon) - F_A(\varepsilon - \omega)]}{(\omega - 2\varepsilon)^2 + 4\gamma^2}, \quad (12.68)$$

where we have performed the integrals assuming the distributions are effectively constant.³ By applying the same approach to $[D_{\psi^\dagger\psi}^{-1}]^K$ one has:

$$2n_\psi(\omega) + 1 = \frac{\kappa(2n_\psi(\omega) + 1) + \frac{ng^2\gamma}{(\omega - 2\varepsilon)^2 + 4\gamma^2} [1 - F_B(\varepsilon)F_A(\varepsilon - \omega)]}{\kappa + \frac{ng^2\gamma}{(\omega - 2\varepsilon)^2 + 4\gamma^2} [F_B(\varepsilon) - F_A(\varepsilon - \omega)]}. \quad (12.69)$$

From this expression one may first note that if $\gamma = 0$ (or more generally if $\kappa \gg g^2\gamma/[(\omega - 2\varepsilon)^2 + 4\gamma^2]$), the the system distribution is the same as the distribution of the decay bath (the photons outside the cavity) and so $n_\psi(\omega) = n_\Psi(\omega)$. On the other hand, if $\kappa = 0$, (or more generally, if $\kappa \ll g^2\gamma/[(\omega - 2\varepsilon)^2 + 4\gamma^2]$, which can occur near $\omega = 2\varepsilon$), the distribution is set by the pumping bath. In this case, the important terms in Eq. (12.69) are:

$$2n_\psi(\omega) + 1 = \frac{1 - F_B(\varepsilon)F_A(\varepsilon - \omega)}{F_B(\varepsilon) - F_A(\varepsilon - \omega)} = \coth \left(\frac{\beta}{2} \left[\varepsilon - \frac{\mu_B}{2} - \varepsilon + \omega - \frac{\mu_B}{2} \right] \right), \quad (12.70)$$

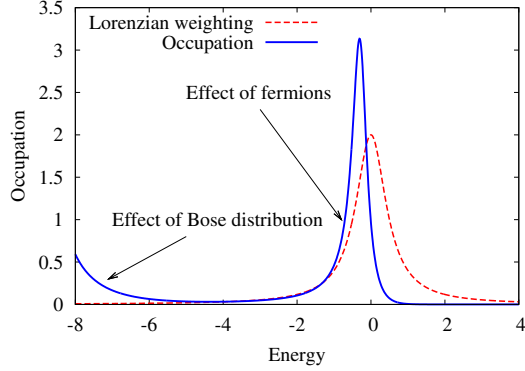
which is a Bose distribution with the temperature and chemical potential of the pumping bath. Thus, the photon distribution interpolates between the decay and pumping bath, depending on the efficiency of coupling as a function of energy. An illustration of how this might look when the chemical potential of the decay bath is not too dissimilar from the pumping bath is shown in Fig. 12.8, however for realistic parameters, the chemical potential of the decay bath should be taken to $-\infty$.

12.6.3 Normal-state instability for a simple laser

As for the mean-field theory, it is instructive to compare the results of Sec. 12.6.2.1 to those for a simple laser, in which pumping tries to fix the inversion of the gain medium, independent of frequency. The instability of the normal state can still be determined by the inverse retarded Green's function, which may in turn be found by the response of Eq. (12.39)–(12.41) to an applied force $Fe^{-i\omega t}$ acting on the photons.

³ Formally, the approximation consists of performing the contour integral, taking into account the poles at $\nu = -\omega + \varepsilon_i + i\gamma$ and $\nu = -\varepsilon + i\gamma$, but neglecting the poles from $F_{A,B}(\nu)$ which are at $\nu = \{-\omega + \mu_B/2, -\mu_B/2\} + i(2n+1)\pi T$, along with neglecting $\beta\gamma$ in evaluating the residues.

Fig. 12.8 Cartoon of occupation function set by competition of bosonic bath and fermionic bath, with effect of fermionic bath moderated by a Lorentzian filter depending on excitonic energy. The chemical potential of the decay bath is at -9 , and that of the pumping bath is just below zero.



If the force is weak, then Eq. (12.41) reduces to $N_i = N_0$, and taking $\lambda_{\perp} = 2\gamma$ as found previously, the equations to solve are:

$$\partial_t \psi = -i\omega_0 \psi - \kappa \psi + \sum_i g_i P_i + F e^{-i\omega t}, \quad \partial_t P_i = -2i\varepsilon_i P_i - 2\gamma P_i + g\psi N_0, \quad (12.71)$$

hence writing the response as $\psi = iD_{\psi^\dagger\psi}^R(\omega) F e^{-i\omega t}$, and eliminating P_i gives:

$$[D_{\psi^\dagger\psi}^R]^{-1}(\omega) = \omega - \omega_0 + i\kappa + \sum_i \frac{g_i^2 N_0}{\omega - 2\varepsilon_i + i2\gamma}. \quad (12.72)$$

As in the mean-field case, this same equation can be recovered from the microscopic non-equilibrium model by taking $F_{A,B}$ to be independent of frequency, and identifying $N_0 = -(F_B - F_A)/2$. The form of the inverse retarded Green's function makes much clearer the implications of this absence of frequency dependence. For the imaginary part of Eq. (12.72) to be zero, it is clearly necessary that $N_0 > 0$, so a region of gain can only exist when inverted.

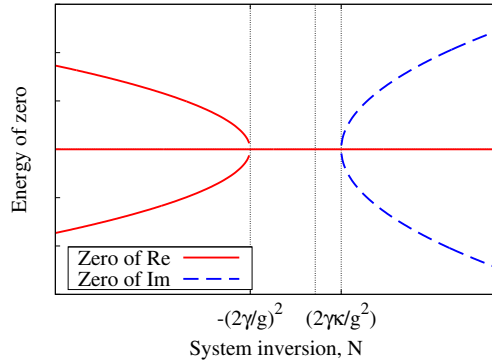
In the special case of $\varepsilon_i = \varepsilon = \omega_0/2$, $g_i = g$, the zeros of the real and imaginary parts can be found explicitly to be

$$\mu_{\text{eff}} = 2\varepsilon \pm \sqrt{g^2 n N_0 \frac{2\gamma}{\kappa} - 4\gamma^2}, \quad \xi = 2\varepsilon, \quad 2\varepsilon \pm \sqrt{-4\gamma^2 - g^2 N_0 n}, \quad (12.73)$$

where n is the number of two-level systems as before. From the zeros of the imaginary part, one sees that a region of gain exists only for $N_0 > 2\kappa\gamma/g^2n$ (note that this is the laser threshold condition discussed in section 12.5.2). On the other hand, a splitting of the zeros of the real part ξ exists only if $N_0 < -4\gamma^2/g^2n$. Thus the instability of the normal state only occurs after the normal mode splitting has collapsed. This is illustrated in Fig. 12.9. In this figure, it is also clear that as soon as there is a region of gain, there is an instability. This is quite different from Fig. 12.7, where a region of gain, and thus zeros of the imaginary part, emerged at a lower pumping strength than was required for the instability. This meant that in the non-

equilibrium condensate, a diverging distribution function exists before condensation occurs, whereas for Fig. 12.9, the distribution function has no divergence in the normal state.⁴

Fig. 12.9 As for Fig. 12.7 but for the results of the Maxwell-Bloch equations, showing the rather different behaviour in the extreme laser limit, plotted for $\omega_0 = 2\varepsilon$.



12.7 Fluctuations of the condensed system

When condensed, the derivation of the spectrum from the inverse Green's function written previously becomes much more involved, but the essential features of the spectrum can be determined by considering the symmetries that the system must possess — the results of this analysis are confirmed by the exact expressions for the inverse Green's functions. In particular, one may combine the Hugenholtz-Pines relation, mentioned at the end of Sec. 12.6.1, with the analytic properties of the Green's functions which imply $[D_{\psi\psi^\dagger}^R]^{-1}(\omega, p) = [D_{\psi^\dagger\psi}^R]^{-1}(-\omega, p)^*$, $[D_{\psi^\dagger\psi^\dagger}^R]^{-1}(\omega, p) = [D_{\psi\psi}^R]^{-1}(-\omega, p)^*$. From these general considerations, one may find that the most general structure for sufficiently small ω, k is:

$$D_{\psi^\dagger\psi}^R(\omega, k) = \frac{C}{\det([D^R]^{-1})} = \frac{C}{\omega^2 + 2i\omega x - c^2 k^2}, \quad (12.74)$$

where x is an effective linewidth, and c an effective sound velocity. The form of this expression is dictated by: the need to combine symmetry under $k \rightarrow -k$; the existence of a finite linewidth; and the pole at $\omega = 0, k = 0$ that is ensured by the

⁴ If one considers the more general case with detuning, $\omega_0 \neq 2\varepsilon$, a region of gain may appear before the instability occurs. Furthermore, if one also has inhomogeneous broadening, $\varepsilon \neq \varepsilon_j$, and different inversion for different two-level systems, a region of gain can coexist with a splitting of the normal states. However, the results for the non-equilibrium condensate shown in Fig. 12.7 had neither detuning nor inhomogeneous broadening; hence in the absence of such complications, the difference between the non-equilibrium condensate and a simple laser are particularly obvious.

Hugenholtz-Pines relation. Higher order contributions could exist (and in fact do exist) for larger ω, k , but the $\omega, k \rightarrow 0$ structure is fixed by these considerations.

The above structure means that the poles of the Green's function for small k are diffusive, i.e. $\omega^* = -ix \pm i\sqrt{x^2 - c^2 k^2}$, meaning that long wavelength excitations decay, but with a lifetime that diverges for one mode as $k \rightarrow 0$. This same form is also recovered from other approaches to non-equilibrium condensates[45], including also the case of a parametrically pumped polariton system[60]. Note that if one were to naively extract a Landau critical velocity from the real part of ω^* , then this critical velocity would vanish. There has been some work on how the concept of the Landau critical velocity may be generalised for parametrically pumped condensates[61, 62, 63], however the full implications of the diffusive structure on superfluidity of incoherently pumped non-equilibrium condensates remains an open question. Because the polariton system is two-dimensional, phase fluctuations can be expected to play a particularly important role, therefore the remainder of this section will discuss how the above form of the Green's function determines the long-time correlations, and hence the lineshape, and how this connects to other approaches to deriving the polariton lineshape.

To take full account of the phase fluctuations, one must reparameterise the fluctuations as $\psi = \sqrt{\rho} + \pi e^{i\phi}$. In order that one works with fields for which there is a macroscopic expectation of $\langle \psi \rangle$ this reparameterisation must be performed in real space, and in terms of the fields on the forward and backward contours, rather than the symmetric and antisymmetric combinations of these fields. (Note that the macroscopic expectation of the anti-symmetric combination ψ_- vanishes [30].) To describe the long-time correlations, we wish to find the first order coherence function (experimental determination of which is discussed in section 11.7 of Chapter 11), which in our formalism is given by $D_{\psi^\dagger \psi}^{fb}(t) = -i\langle T_c[\psi(t, f)\psi^\dagger(0, b)] \rangle$, and corresponds to the Fourier transform of the luminescence spectrum, $\mathcal{L}(\omega)$. Since it is the phase fluctuations that dominate the long time behaviour, one may write this asymptotic behaviour in the form:

$$D_{\psi^\dagger \psi}^{fb}(t) \simeq \rho_{QC} \langle \exp[i(\phi(t) - \phi(0))] \rangle = \rho_{QC} \exp[-f(t)], \quad (12.75)$$

where ρ_{QC} is the quasi-condensate density. The function $f(t)$ is given by the phase-phase correlation functions, and in two dimensions is given by:

$$f(t) = i \left[D_{\phi\phi}^{fb}(t) - D_{\phi\phi}^{fb}(0) \right] = \int \frac{d\omega}{2\pi} \int \frac{kd k}{2\pi} [1 - e^{-i\omega t}] i D_{\phi\phi}^{fb}(\omega, k). \quad (12.76)$$

Note that expressions (12.75) and (12.76) are determined by taking the phase fluctuations to all orders. The density fluctuations give no time dependence at long times, their effect appears only in the difference between the quasi-condensate density ρ_{QC} and the total density ρ .

Since $D_{\phi\phi}^{fb}$ corresponds to the luminescence spectrum, its relation to Keldysh and retarded Green's functions is as in Eq. (12.62). Assuming that the condensation arises due to pumping, then as in Sec. 12.6.2, the frequency dependence near the ef-

fective chemical potential arises from the behaviour of the inverse retarded Green's function — the frequency dependence of the inverse Keldysh Green's function has no particular singularities near this point. In this case (which is also what is found from the full calculations of the microscopic theory), the singular behaviour of the $D_{\phi\phi}^{fb}$ is given by $D_{\phi\phi}^{fb} \sim |D_{\phi\phi}^R|^2$, and so:

$$f(t) = \int \frac{d\omega}{2\pi} \int \frac{kdk}{2\pi} \frac{(C^2/\rho)}{|\omega^2 + 2i\omega x - c^2k^2|^2} [1 - e^{-i\omega t}]. \quad (12.77)$$

(The factor of $1/\rho$ occurs from the relation of phase-phase Green's functions to $\psi, \psi^\dagger$ Green's functions). As one expects for a two-dimensional system, after integrating over ω , the above integral reduces to an expression $\sim \int dk/k$, and so one has logarithmic behaviour, cut off at high k by a maximum energy of excited modes, and at small k by the time dependence. At small k , the poles of the ω integral are at $\omega = \pm 2ix, \pm i(ck)^2/2x$; the first of these has a finite residue as $k \rightarrow 0$, while the latter has a residue that is diverging, and thus dominates the behaviour. The asymptotic behaviour is thus given by:

$$f(t) = \int \frac{kdk}{2\pi} \frac{C'}{4x(ck)^2} [1 - e^{-c^2k^2t/2x}] \quad (12.78)$$

(where C' is a new constant). This expression has a cutoff for small k given by $k \sim \sqrt{x/t}/c$; thus one still has power law correlations as for an equilibrium two dimensional gas, but with a different power law, now set not only by the condensate density but also by the pumping and decay strength. Were one to calculate also the long-distance correlations at equal times, one would note another difference from equilibrium. In equilibrium, the decay of long-time equal-position correlations, and long-distance equal-time correlations have the same power laws. For the spectrum in Eq. (12.74), the power-law for long-distance equal-time correlations is twice that of long-time equal-position decay. This is because the low momentum cutoff for long distances is always $k \sim 1/r$, whereas the long-time cutoff is $k \sim 1/ct$ in equilibrium, but $k \sim \sqrt{x/t}$ here.

12.7.1 Finite-size effects – lineshape of trapped system

For a confined system, the integral over k modes is replaced by a sum over a discrete set of modes; i.e.:

$$f(t) = \sum_n \int \frac{d\omega}{2\pi} \frac{C' [1 - e^{-i\omega t}]}{|\omega^2 + 2i\omega x - \xi_n^2|^2} \simeq \sum_n \frac{C'}{4x\xi_n^2} [1 - e^{-\xi_n^2 t/2x}]. \quad (12.79)$$

In this form, one may then consider how the value of the sum depends on the relative size of the mode spacing ΔE , the low energy cutoff $\sqrt{x/t}$, and the maximum energy

$E_{\max}$. Let us assume the maximum energy is large, then we have a picture something like Fig. 12.10.

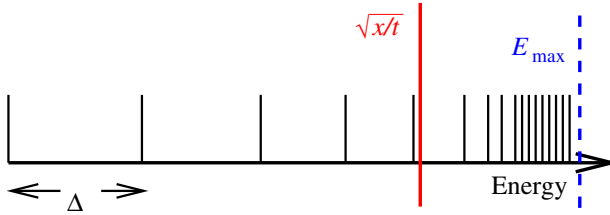


Fig. 12.10 Spacing of discrete energy levels, and upper/lower cutoff energies

The sum can be divided into parts above and below the low energy cutoff, giving:

$$f(t) \simeq C' \left[\sum_{n=0}^{\xi_n < \sqrt{x/t}} \frac{t}{8x^2} + \sum_{\xi_n > \sqrt{x/t}}^{\xi_n = E_{\max}} \frac{1}{4x\xi_n^2} \right] \quad (12.80)$$

If both of these sums have many terms, then they may be approximated by integrals, and if the density of states $\nu(\xi)$ is $\nu_0\xi$ as it would be for a two-dimensional system with $\xi_n = cp_n$ (as illustrated in Fig. 12.10) then this becomes:

$$f(t) \simeq C' \nu_0 \left[\int_0^{\sqrt{x/t}} \frac{t}{8x^2} \xi d\xi + \int_{\sqrt{x/t}}^{E_{\max}} \frac{\xi d\xi}{4x\xi^2} \right] \simeq \frac{C' \nu_0}{16x} + \frac{C' \nu_0}{4x} \ln \left(E_{\max} \sqrt{\frac{t}{x}} \right). \quad (12.81)$$

What is to be noted here is that the number of terms in the first part compensates the t dependence, leading to a harmless constant. If however the number of terms in the first term is small, or is in fact truncated at its minimum value of one (which will inevitably occur for large enough t), then one instead has something of the form:

$$f(t) \simeq C' \left[\frac{t}{8x^2} + \frac{\nu_0}{4x} \ln \left(\frac{E_{\max}}{\Delta E} \right) \right], \quad (12.82)$$

and so one has exponential decay of coherence at long times, arising from the restricted number of modes.

If the mode spacing is such that only a single mode is involved, then Eq. (12.79) reduces to a single term in the sum, $\xi_0 = 0$, and the result becomes very similar to the form found from models of phase noise for a single mode condensate [64, 65], for which the lineshape interpolates between Gaussian and Lorentzian:

$$f(t) = C' \int \frac{d\omega}{2\pi} \frac{1 - e^{-i\omega t}}{(\omega^2 + 0^2)(\omega^2 + 4x^2)} = \frac{C'}{16x^2} [2xt - 1 + e^{-2xt}], \quad (12.83)$$

hence the decay of coherence varies from t^2 at short time (giving Gaussian lineshape at high frequencies) to t at long times (giving a Lorentzian peak at low frequencies). This result is exactly as one expects for phase noise from varying densities[65]:

$$\partial_t \phi = -iUN, \quad \partial_t N = -\Gamma N + F_\Gamma(t), \quad (12.84)$$

where F_Γ is a Gaussian delta correlated noise noise noise with strength P_Γ . Solving these equations in Fourier space, one has:

$$\langle |\phi_\omega|^2 \rangle = \frac{U^2}{\omega^2} \langle |N_\omega|^2 \rangle = \frac{U^2 P_\Gamma}{\omega^2 (\omega^2 + \Gamma^2)}, \quad (12.85)$$

which is the same form as in Eq. (12.83)

This section thus shows another distinction between condensates and lasers in terms of many-mode or single mode fluctuations. If the system is large the spatial fluctuations resulting from the continuum of modes give rise to (in two dimensions) a power-law decay of correlations as for an infinite, equilibrium, two-dimensional quasi-condensate. For smaller systems, or at longer times, the power law crosses over to exponential decay, given by a fluctuations within the single lowest energy mode (the other modes are too high in energy to be relevant), as is characteristic for lasers.

12.8 Summary

This chapter has discussed in detail how the non-equilibrium Green's function formalism can be applied to study a model of microcavity polaritons, driven out of equilibrium by coupling to two baths. This model system, while not incorporating all features of the real system, allows one to make particularly transparent connections between laser theory and equilibrium descriptions, as well as allowing clear illustrations of the consequences of the approximations typically used for simple lasers. By considering steady states of the system in which there is a coherent photon field, one finds a criterion for condensation to occur, and can find a self-consistency condition which determines how the amplitude and frequency (effective chemical potential) of the coherent field depend on the strength of the pumping and decay. By considering fluctuations about steady states, one can determine whether a given steady state is stable, find the spectrum of possible excitations, and find how this spectrum is populated.

Starting from the normal state, without a condensate, and increasing pumping strength, one finds that fluctuations about the normal state become unstable at the same point that a condensed solution appears. The scenario by which this instability occurs on increasing pumping strength is quite instructive. As pumping strength increases, a region of energies for which there is gain appears in the spectrum. The energy dividing this region of gain from regions of loss defines an effective chemical potential, at which the non-equilibrium distribution function diverges. Instability

occurs at a higher pumping strength, when this effective chemical potential (and thus the region of net gain) reach the normal modes of the strongly coupled system, at which point polariton condensation occurs. Such a description unites the lasing picture of gain exceeding loss with the equilibrium picture of the chemical potential reaching the bottom of the band.

While the above description allows polariton condensation to be discussed in the language of laser theory, the results are rather different from the normal limits assumed for a simple laser theory. However, simpler laser theory results can be recovered within the model discussed here, as corresponding to a high temperature limit. In this high temperature limit, pumping corresponds to effectively white noise, and this was shown to mean that gain only exists when pumping bath is inverted. This has the consequence that in this high temperature limit, lasing and strong coupling do not coexist, whereas they can in the low temperature polariton condensate.

When considering fluctuations about the condensed state, a somewhat different distinction between simple lasers and the polariton condensate emerges: the effect of finite system size, and the spectrum of collective phase modes. For an infinite two dimensional system, the decay of coherence at long distances and long times is power law, as in equilibrium (but with different powers). For a finite system, the effects of finite lifetime and finite size combine to lead to exponential decay at long times. In the limits of very small system size, the standard result for phase noise in a single mode condensate is naturally recovered.

To summarise, the approach presented here provides a way to connect a number of different approaches to equilibrium and non-equilibrium condensates, as well as theories of lasers, in a transparent manner, allowing one to understand the significance of various approximations, as well as the relations between some of the other approaches one may use.

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Index

Abrikosov-Gorkov approach, 295

Absorption

- absorption coefficient, 168
- absorption lineshape, 19
- absorption spectroscopy, 91
- absorption spectrum, 144, 189
- magneto-optical absorption, 12, 172
- resonant absorption, 159

adiabatic elimination, 71

adiabatic pulse, 70

Aharonov-Bohm phase, 172

Anderson's theorem, 295

anti-crossing, 13, 135

anti-dots, 182

anti-trapping potential, 274

atom-cavity system, 216

atomic force microscopy (AFM), 42

Auger effect, 10

ballistic expansion, 274

Bell's theorem, 216

Bloch function, 68

Bloch sphere, 64, 65, 119, 121, 123

Bogolyubov approach, 196

Bohr magneton, 69

Boltzmann equation, 269

Born approximation, 302

Born-Markov approximation, 231

Bose statistics

- Bose distribution function, 22
- Bose gas, 183, 266
- Bose glass, 210, 276
- Bose statistics, 231
- Bose-Einstein condensation (BEC), 208
- Bose-Hubbard model, 210
- bosonic operator, 195
- bosonic stimulation, 269

bosonic systems, 319

Bosonization procedure, 234

weakly interacting bosons, 294

Bragg condition, 186

Broadening

- homogeneous broadening, 176
- inhomogeneous broadening, 17, 245
- tunneling-assisted broadening, 33

C-numbers, 246

capping, 43

carrier capture, 49

central limit theorem, 28

chemical potential, 198, 298

chiral media, 153

chirality, 152

cholesteric liquid crystals, 171

circular birefringence, 152

circular dichroism, 88, 152

co-circular channel, 252

Coherence

- coherence parameter, 206
- coherence time, 17
- coherent control, 217
- coherent excitation, 93
- coherent manipulation, 25
- coherent Maxwell-pseudospin formalism, 159
- coherent population trapping (CPT), 65, 76, 178
- coherent spin dynamics, 88
- coherent superposition of states, 146
- mode-locked spin coherence, 104
- phase coherence, 285
- photon coherence parameter, 196
- spatial coherence, 284
- two-exciton pair coherences, 228

coincidence detection, 235
 cold atomic gases, 294
 colloidal nanocrystals, 41
 complex propagation factor, 168

Condensation

BCS-BEC crossover theory, 307
 condensate emission spectrum, 277
 condensate linewidth, 277
 critical density, 286
 critical temperature, 297
 critical temperature for polariton condensation, 182
 metastable quantum condensates, 294
 multimode condensate, 277
 non-equilibrium condensation, 294
 polariton Bose-Einstein condensation (BEC), 182, 267
 spinor condensate, 282
 control pulse, 78

Correlation

anomalous correlations, 312
 correlation time, 28
 correlations in momentum space, 274
 Coulomb correlation, 217
 first order time correlations, 285
 four-particle correlation term, 225
 long range correlations, 277
 multi-time correlation functions, 239
 quantum correlations, 235, 243, 251
 spatial correlations, 276
 spatial first order correlations, 283
 counter-circular channel, 252
 Courant condition, 163
 cyclotron energy, 12

D'yakonov-Perel mechanism, 48

DCTS-Langevin approach, 254

decay bath, 298

decoherence, 12, 295

deep level transient spectroscopy, 34

deformation potential, 254

density matrix, 76

Dephasing

ensemble spin dephasing time T_2^* ,
 excitation induced dephasing, 228
 extrinsic dephasing mechanism, 26
 pure dephasing, 10
 spin decoherence time, T_2 , 48

Di Vincenzo criteria, 64, 86

diamagnetic shift, 13, 135

diffusion coefficients, 238

Dipole

dipole matrix element, 97
 dipole moment, 159

dipole-coupling perturbation, 159
 dipole-dipole interactions of nuclear spins,
 54
 optical dipole matrix element, 158

Disorder

disorder coupling constants, 188
 non-pair-breaking disorder, 295
 oscillator strength disorder, 187
 pair-breaking disorder, 295
 photonic disorder, 276
 positional disorder, 187
 propagator of disordered system, 189
 static disorder, 295
 strong disorder, 203
 weak disorder, 203
 dynamics controlled truncation scheme (DCTS), 223
 Dyson equation, 300

effective coupling, 205

effective dielectric constant, 161

effective g factor, 69

effective gain, 273

effective ionization energy, 34

effective magnetic field, 149

effective mass method, 158

effective medium approximation, 157

effective nuclear field, 99

effective refractive index, 163

eight-shaped curve, 241

Einstein equations, 236

electrical injection, 46

electrical injection from magnetic contacts, 46

electromagnetically-induced transparency (EIT), 82, 178

electron spin resonance (ESR), 66

Elliott-Yafet mechanism, 48

ellipticity, 78, 154

enantiomer, 156

Entanglement

entangled idler states, 244
 entanglement, 14, 251
 Entanglement of Formation (EOF), 252
 exciton Bohr radius, 227
 many-particle entangled quantum states, 216
 polariton entanglement, 256
 polarization-entangled photon pairs, 230
 polarization-entangled quantum state, 230
 envelope function, 68
 equivalent isotropic dielectric function, 158
 evolution operator, 74

Exciton

exciton Aharonov-Bohm effect, 201
 exciton detuning, 204

- exciton envelope function, 222
- exciton g-factor, 49
- exciton insulator, 197
- exciton inter-dot transfer amplitudes, 193
- exciton localization length, 187
- exciton operators, 185
- exciton spin-flip processes, 196
- exciton superfluid order parameter, 196
- exciton transfer energy, 198
- excitonic gain medium, 295
- Frenkel exciton, 223
- neutral exciton, 33
- radiative lifetime of excitons, 53
- Excitonic states**
 - biexciton, 134, 206, 221
 - bright and dark exciton states, 49, 143
 - bright-dark exciton splitting, 93
 - charged excitons, 43
 - exciton complexes, 43
 - ferromagnetic exciton ground state, 197
 - fine structure splitting, 44
 - paramagnetic exciton ground state, 197
 - trion, 49, 67
 - trion energy, 191
 - trion radiative recombination, 99
 - trion singlet state, 99
- Förster transfer, 195
- Far InfraRed, 14
- Faraday**
 - Faraday configuration, 49, 146
 - Faraday effect, 152
 - Faraday rotation, 47
- fermionic operator, 195, 297
- Feynman rules, 314
- fidelity, 65, 80, 194
- field-effect heterostructure, 32
- field-induced tunneling, 34
- Finite-Difference Time-Domain (FDTD)
 - technique, 162
- Fluctuation**
 - energy fluctuations, 187
 - extrinsic reservoir fluctuation dynamics, 32
 - fluctuation-dissipation theorem, 236
 - number fluctuations, 285
 - phase fluctuations, 323
 - quantum fluctuations, 183, 229
 - quantum well thickness fluctuations, 275
 - shot-noise fluctuations, 286
- flux quantum, 172
- flying qubit, 67
- Fock space, 219
- Four Wave Mixing**
 - Four Wave Mixing, 17
 - non-degenerate four-wave mixing, 230
 - transient (frequency-resolved) four-wave mixing (FWM), 229
- four-level quantum system, 174
- four-particle kernel, 227
- Fourier-transform spectroscopy, 30
- frequency focusing, 113
- frequency synchronization, 277
- g-factor tensor, 93
- gain coefficient, 168
- Gate**
 - conditional phase gate, 193
 - gate errors, 203
 - ideal gate matrix, 194
 - logic gate, 119
 - quantum gate, 55
 - real gate matrix, 194
 - surface gate structures, 41
 - universal two-qubit gate, 193
- gauge transformation, 303
- Gaussian stochastic process, 28
- Gell-Mann's generators, 160
- Ginzburg-Landau equation (CGLE), 273
- Green**
 - advanced Green's function, 299
 - non-equilibrium Green's functions
 - approach, 296
 - photon Green's function, 312
 - retarded Green's function, 299
- Gross-Pitaevskii equation, 269, 296
- Hanbury Brown-Twiss interferometer, 286
- Hartee-Fock (mean-field) term, 227
- Heisenberg**
 - Heisenberg equation, 301
 - Heisenberg equations, 226
 - Heisenberg picture, 224
 - Heisenberg-Langevin approach, 235
 - Heisenberg-Langevin equations, 296
 - Heisenberg-type exchange interaction, 132
- Hole**
 - heavy hole, 68, 221
 - heavy-hole light-hole splitting, 191
 - light hole, 68, 221
 - split-off hole band, 68
- Hopfield coefficients, 189
- Huang-Rhys term, 18
- Hubbard**
 - generalized Hubbard operators, 223
 - Hubbard Hamiltonian, 196
 - Hubbard model, 206
- Hugenholtz-Pines relation, 315
- hyperbolic secant pulse, 70

incoherent dynamics, 235
indium-flush technique, 46

Interaction

anharmonic coupling, 10
anisotropic exchange, 95
asymmetric exchange interaction, 49
Coulomb interaction, 69
electron-electron interaction, 219
electron-hole exchange interaction, 44, 91
electron-phonon interaction, 14, 234
electron-photon matrix element, 158
exchange interaction, 48
exciton-exciton interaction, 195
hole-hole interaction, 219
hyperfine interaction, 41, 48, 86
interacting polaritons, 231
interaction Hamiltonian, 223
interaction picture, 66, 301
long-range coupling, 191
quadrupolar interaction, 53
screened Coulomb interaction, 219
screening, 295

Interband

interband dipole matrix element, 222
interband excitation, 10
interband optical transitions, 10
interband relaxation, 165
interband transitions, 18

interface roughness, 245

interferogram, 30

intermixing, 43

Intraband

intraband absorption, 11
intraband optical transitions, 10
intraband relaxation, 165

intrinsic anharmonicity, 17

intrinsic radiative limit, 32

Ising term, 135

isoelectronic impurity, 132

Josephson currents, 277

Jump

asymmetric jump event, 29
jump processes, 27
quantum jump approach, 194
symmetric jump processes, 29

Keldysh

Keldysh block, 305
Keldysh Green's function, 296
Keldysh non-equilibrium diagrammatic technique, 298

Knight field, 116

Kubo theory, 27

ladder operators, 135

Larmor frequency, 87

Laser

atom lasers, 295
free electron laser, 15
laser coherence, 208
laser frame, 66
laser gain medium, 165
laser threshold condition, 309
mode-locked laser, 88
polariton lasing, 270
quantum cascade laser (QCL), 16
random lasers, 295
vertical cavity surface emitting laser (VCSEL), 270

lattice temperature, 258

level shift operator, 192

light field quantization, 229

light-emitting diode (spin-LED) structures, 46

linear dichroism, 88

linear dynamics, 224

lineshape of trapped system, 325

Liouville-von Neumann equation, 70, 135

local dielectric tensor, 157

macro-atom model, 13

macroscopic magnetization, 89

magic-angle, 240

magnetic circular dichroism (MCD), 153

magnetic impurities, 132

magnetic ion, 132

magnetization inversion, 66

magneto-chiral optical effect, 152, 178

magons, 294

Markov

Markov approximation, 236, 296

Markov chain, 27

Markovian modulation, 27

non-Markovian process, 227

Maxwell's curl equations, 161

Maxwell-Bloch equations, 308

Maxwell-Boltzmann distribution, 270

Maxwell-pseudospin equations, 157

mean-field approach, 148, 296

memory kernel, 227

mesa structures, 182

metal-insulator transition, 172

metallic nano-particles, 260

metamaterials, 153

Michelson interferometer, 30

Microcavity

Arsenide-based microcavity, 267

cavity exciton-polaritons, 216

cavity photon mass, 186
 cavity quality factor Q , 198
 cavity quantum electrodynamics (QED), 216
 planar microcavity, 182
 polariton amplification, 227, 247
 polariton basis, 229
 polarization-selective cavity, 196
 semiconductor microcavities (SMCs), 216
 Telluride-based microcavity, 267
 two-dimensional array of cavities, 183
Microcavity polaritons
 Bragg cavity polariton, 186
 central polariton (CP), 199
 exciton-photon coupling constant, 185
 exciton-polariton, 182
 longitudinal-transverse splitting, 222
 lower polariton (LP), 199
 microcavity polaritons, 294
 polariton effective mass, 182
 polariton Hamiltonian, 185
 polariton linewidth, 194
 polariton mixing, 186
 polariton modes, 182
 polariton splitting, 201
 polartion lasing, 267
 quantum well polaritons, 200
 spin-entangled cavity polaritons, 249
 strong coupling regime, 216, 266
 upper polariton (UP), 199
 microdisks, 288
 mixed states, 49
 mode switching, 118
 Molecular Beam Epitaxy, 64
 molecular transitions, 159
 momentum conservation, 190
 Monte Carlo simulation, 196
 motional narrowing, 28, 276
 Mott lobes, 206
 multiple band models, 44
 nanocavity, 183
Nanotube
 achiral nanotubes, 154
 armchair nanotubes, 154
 chiral indices, 159
 dielectric polarizability of a single carbon nanotube, 158
 electronic band structure of a SWCNT, 155
 single-wall carbon nanotubes (SWCNTs), 153
 zig-zag nanotubes, 154
 natural optical activity, 152
 negative refractive index, 153

Noise

coloured noise, 309
 Gaussian stochastic noise, 28
 Langevin noise source operators, 236
 operator noise sources, 236
 quantum Langevin forces, 246
 quantum Langevin noise approach, 236
 random telegraph (noise), 27
 stochastic noise terms, 246
 white noise, 309
 non-classical states, 217
 non-equilibrium diagrammatic approach, 296
 non-equilibrium path integral approach, 296
 non-equilibrium system, 294
 non-resonant excitation, 49
 nonlinear coherent regime, 153
 nonlinear dynamics, 217, 226
 nonperturbative dynamics, 231
 normal-state instability, 321
 nuclear magnetic resonance, 28
 open quantum systems, 216
 optical activity, 152
 optical gyrotropy, 153
 optical injection, 46
 optical orientation, 88
 optical selection rules, 47, 69
 optically active states, 93
 order parameter, 197, 206
Oscillator
 coupled oscillators model, 152
 damped oscillation, 97
 single-oscillator model, 152
 Overhauser field, 53

Parametric processes

mixed parametric processes, 255
 parametric down-conversion, 243
 parametric emission, 226
 parametric oscillation scheme, 267
 parametric scattering, 226
 pattern formation, 273
 Pauli spin blocking, 43, 49, 100
 Penrose-Onsager criterion, 283
 periodic boundary conditions, 201
 perturbation theory, 71

Phase

equilibrium phase transition, 294
 phase boundary, 310
 phase delay, 170
 phase diagrams, 198
 phase dynamics, 227
 phase locking, 197
 phase of order parameter, 272

- phase shift, 176
- phase space filling, 233
- phase synchronization condition, 102
- quantum phase, 183
- quantum phase transition (QPT), 183
- superfluid phase, 208
- Phonon**
 - acoustic phonons, 15, 17
 - deformation potential, 18
 - Fröhlich interaction, 13, 15
 - Fröhlich matrix element, 15
 - LO-phonon relaxation, 269
 - optical phonons, 13
 - phonon absorption, 18
 - phonon bath, 18
 - phonon bottleneck effect, 12
 - phonon emission, 18
 - phonon sidebands, 17
 - phonon-assisted tunneling, 34
 - phonons, 12
 - piezo-electric field, 11
 - polaron, 13
 - two-phonon processes, 21
 - zero phonon level, 13
 - zero phonon line (ZPL), 19
- phonon bottleneck effect, 112, 240
- photocurrent spectroscopy, 33, 44
- Photoluminescence**
 - cw PL spectroscopy, 49
 - luminescence lifetime, 49
 - micro-photoluminescence, 276
 - micro-photoluminescence (PL), 43
 - parametric luminescence, 234
 - photoluminescence, 26
 - photoluminescence intensity, 315
 - photoluminescence spectroscopy, 91
 - time-resolved photoluminescence, 49
- Photon**
 - photon anti-bunching, 287
 - photon bunching, 286
 - photon hopping, 195
 - photon insulator, 197
 - photon lasing, 267
 - photon pair generation, 25
 - photon superfluidity, 206
 - photon transfer energy, 198
 - photon-counting coincidence detections, 241
 - photon-exciton coupling function, 193
- photonic crystal, 183
- photonic metamaterials, 171
- point-dipole crystal, 182
- Polarisation**
 - electronic polarization, 221
 - exciton polarization, 191
 - macroscopic polarisation, 162
 - negative circular polarization (NCP), 50
 - polarization-mixing, 226
 - spin polarization, 99
- post-processing, 252
- projection operator, 192
- pseudo-potential method, 10
- pseudopotential models, 44
- pulse area, 97, 163
- pulse reshaping, 165
- Pump-Probe**
 - pump-probe experiments, 15
 - pump-probe setup, 144
 - time-resolved pump-probe Faraday technique, 88
- Pumping**
 - co-circular polarized pumps, 107
 - counter-circular polarized pumps, 107
 - pump protocol, 107
 - pump-induced polariton energies renormalization, 239
 - pumping, 295
 - pumping bath, 298
 - selective optical pumping, 133
 - two-pump scheme, 247
- pure state, 194
- quantum beat spectroscopy, 91
- quantum complementarity, 216, 243
- quantum degenerate regime, 266
- Quantum Dot**
 - charged QDs, 49
 - charged quantum dots, 89
 - composition of QDs, 44
 - doped QDs, 49
 - double QDs, 133
 - electrostatically defined QDs, 41
 - lateral coupling of QDs, 46
 - lattice constant mismatch, 41
 - lattice of charged quantum dots, 182
 - lattice of quantum dots in a planar microcavity, 182
 - lithography defined QDs, 41
 - modulation doping, 12
 - neutral dots, 98
 - post growth annealing, 44
 - QD molecule, 55
 - quantum dot (QD), 10
 - quantum dot ensembles, 86
 - relaxation in QDs, 12
 - self-assembled quantum dots, 10
 - single QD, 55
 - single QD spectroscopy, 44

- stacked quantum dots, 16
- Stranski-Krastanov growth mechanism, 41
- three dimensional arrays of QDs, 182
- transport quantum dots, 64
- tunnel-coupled QDs, 72
- vertically-coupled QDs, 46
- quantum Hall bilayers, 294
- quantum information processing (QIP), 40
- quantum kinetic equations, 296
- quantum nondemolition measurements, 216
- quantum regression theorem, 296
- quantum simulators, 183
- quantum state tomography, 257
- Quantum well**
 - coupled quantum wells, 197
 - periodic quantum well Bragg structures, 182
 - quantum well, 14
- quasi-mode approximation, 222
- quasi-mode model, 191
- quasiangular momentum, 158
- quasistatic approximation, 158
- qubit, 25, 40
- Rabi**
 - exchange-induced Rabi oscillations, 145
 - Rabi flop, 120
 - Rabi frequency, 137
 - Rabi oscillations, 97, 137
 - Rabi problem, 70
 - Rabi splitting, 276
- radiative coupling, 195
- radiative lifetime, 159
- real state vector, 159
- recombination, 70
- relaxation bottleneck, 267
- relaxation function, 27
- resonant amplification, 159
- Reststrahlen band, 15
- rotating frame, 72
- rotating wave approximation (RWA), 73, 162, 222, 248
- Rotation**
 - composite rotations, 65
 - Faraday rotation, 47
 - Faraday rotation bursts, 106
 - Kerr rotation, 47
 - magneto-optical rotatory dispersion (MORD), 153
 - optical rotation angle, 168
 - rotatory power, 152
 - single-shot rotation, 66
 - specific rotatory power, 154
- saturation, 217
- scalability, 45
- scanning tunneling microscopy (STM), 44
- Scattering**
 - anomalous scattering, 284
 - hyper-Raman scattering (HRS), 229
 - incoherent scattering, 239
 - polariton-exciton scattering, 266
 - polariton-polariton interactions, 243
 - polariton-polariton scattering, 266
 - Resonant Rayleigh Scattering, 253
 - two-photon Raman scattering, 229
- Schawlow-Townes linewidth, 285
- Schottky contact, 32
- Schottky diode structure, 43
- sculptured thin films, 171
- second quantization, 184
- secular equation, 220
- self energies, 313
- self energy diagrams, 314
- self-induced transparency, 73
- semiclassical approach, 157
- Semiconductor**
 - diluted magnetic semiconductor, 132
 - direct band gap semiconductors, 68
 - magnetic semiconductor, 46
 - zincblende semiconductors, 86
- signal-idler pair, 241
- single electron transistor, 132
- single mode regime, 118
- single particle scattering, 190
- single photon sources, 25
- single-particle states, 219
- Slater-determinant, 218
- slow modulation limit, 28
- slowly-varying wave approximation (SVEA), 162
- spectral diffusion, 26
- spectral weight, 316
- Spin**
 - all-optical spin echo, 124
 - effective spin-Hamiltonian, 93
 - electron spin, 54
 - electron spin coherence time, 104
 - hole spin, 54
 - longitudinal spin relaxation time, T_1 , 48
 - multi-spin coupling constants, 193
 - nuclear spin precession time, 53
 - nuclear spin-flip rate, 112
 - optical spin control, 70
 - pseudospin, 64
 - pseudospin vector, 74
 - spin basis, 192
 - spin beats, 101
 - spin coherence, 41

- spin coupling, 194
- spin decoherence, 41
- spin decoherence time, 160
- spin energy, 205
- spin flip-flop process, 51
- spin lifetime, 49
- spin memory, 54
- spin multiplet energies, 205
- spin precession, 71
- spin relaxation, 41
- spin singlet state, 50
- spin splitting, 89
- spin synchronization, 102
- spin vector, 65
- spin-B interaction, 172
- spin-orbit coupling, 51, 218
- spinor order parameter, 282
- spintronics, 132
- spontaneous emission, 70, 159
- Stark**
 - optical Stark effect, 65
 - quantum confined Stark effect, 26
 - Stark shift, 27, 43, 203
- stimulation threshold, 269
- STIRAP (Stimulated Raman Adiabatic Passage), 71
- strain interactions, 46
- strain-induced intermixing, 46
- strong localization, 184
- subbands, 219
- superfluid Helium, 294
- superfluid system, 206
- superfluid velocity, 272, 279
- superposition of states, 64
- surface plasmon modes, 260
- switching protocol, 141

- T-matrix, 190
- tensor of the structure constants, 159
- thermalisation, 294
- thermally stimulated escape, 18
- thermo-ionization, 34
- third-order nonlinear susceptibility, 230
- third-order nonlinearity, 17
- three-level model, 140
- three-level system, 74
- tight-binding method, 10, 154
- time evolution operator, 194
- time-dependent Schrödinger equation, 70
- time-ordered product, 300
- time-resolved visibility, 235
- tomographic reconstruction, 252
- torque vector, 159
- transition operators, 232
- transmission electron microscopy (TEM), 44
- transmission spectrum, 167
- truncated Wigner approximation, 296
- two-dimensional electron gas (2DEG), 41
- two-level system, 17
- two-particle states, 227
- two-pulse photon echo, 16
- two-time coincidence, 253
- two-times correlation functions, 252

- ultrafast optical pulses, 55
- ultrafast transient response, 167
- umklapp-processes, 185
- uniaxial stress, 44
- unitary basis transformation, 231
- unitary matrix, 75
- unitary transformation, 64, 303
- universal quantum logic, 75

- vacuum state, 218
- virtual transitions, 18
- visibility, 246
- Voigt configuration, 49, 87, 146
- Vortex**
 - half-quantized vortices, 283
 - quantized vortices, 279
 - vortex, 279
 - vortex string (soliton), 283
 - vortex-antivortex pairs, 280
 - vorticity, 279

- weak coupling regime, 272
- wetting layer, 10, 41
- which-way information, 243
- Wick factorization, 257
- Wiener-Khinchin identity, 283
- Wigner representation, 285
- Wigner transformation, 296

- X-ray diffraction, 44

- Zeeman splitting, 43, 91, 147, 172