Quantum Optics in Semiconductor Microcavities

Portolan Stefano

Tutore
Prof. Rossi Fausto
Dott. Savasta Salvatore

Coordinatore del corso di dottorato
Prof. Mazzetti Piero

Gennaio 2007
agli omini verdi ...

...  
Vai lá menino
Mostra o que o mestre ensinou
Mostra que arrancaram a planta
Mas a semente brotou
E se for bem cultivada
Vai dar bom fruto e bela flor.
Iê, viva Pastinha
Notes and Remarks

This thesis was defended on the 15th of January 2007. The last months we needed to rush in order to complete some simulations, to submit the relevant papers and to write a proper thesis manuscript. The present version presents some refinements we have made throughout the refereeing process we have been experiencing along the subsequent year: some typing errors have been corrected and some little changes in the exposition have been performed in order to clarify some points and get a clearer document.

I really do hope any reader will enjoy the exposition and will get a fresh look into the subject of my (hard) work of these years.

Yours Faithfully

Stefano Portolan

Lausanne, February 2008
Acknowledgements

Sfogliare questa tesi è per me fonte di tanti ricordi. Questi tre anni di dottorato di ricerca sono stati una grande faticata, in tutti i sensi, ed adesso che sono arrivato alla fine e vedo la meta sono più cosciente che mai che non sarei mai riuscito ad ottenere neanche la più piccola parte di tutto questo senza tutto l’aiuto che ho ricevuto dalle persone che mi sono state accanto in questa lunga marcia.

Per prima cosa vorrei ringraziare la mia famiglia. La mia mamma che così tante cose fa sempre per me, penso che alle volte non si renda più neanche conto di quanto metta in secondo piano se stessa per pensare prima a noi, io di mio me ne accorgo eccome e più volte mi sono sentito anche un po' in colpa ad essere così contento di tutta questa attenzione. Il mio papà per quanto ha partecipato in maniera attiva a formarmi come persona (a formarci), indipendentemente da dove si trovasse geograficamente. Vorrei ringraziare anche mia sorella che riesce ad essere vicina e presente anche con tanti km di mezzo. Per ultimo solo per necessità di catalogazione c’è il mio gemello che anche se fisicamente lontanto mi è sempre vicino per tante cose ed in tanti modi (viva MSN!).

Un caro ringraziamento lo vorrei esprimere verso il prof. Fausto Rossi, per avermi accettato all’interno del suo gruppo di ricerca ed insegnato pressoché tutto quello che so. In tutto sono stati quattro anni vissuti per me molto intensamente, con tanti bei ricordi ed emozioni. Confesso che a guardarmi indietro non sono sempre stato all’altezza delle situazioni, ma bisogna pur imparare (anche ad essere adulti).
Ho imparato molto a livello professionale, ma soprattutto sono cresciuto a livello personale assistendo a molte lezioni di vita che si imprimeranno come importanti ricordi nella mia memoria e spero continueranno ad indicarmi la strada anche adesso che dovrò per forza di cose imparare a navigare da solo.

Nel dott. Salvatore Savasta ho incontrato un ottimo fisico e una persona cara e onesta. Mi ha insegnato tante cose, tra fisica e dintorni, sotto la sua supervisione ho lavorato benissimo e spero di riuscire a collaborare con lui ancora e tanto. Se più avanti mi potrò guardare indietro e riconoscere nelle mie azioni un po’ della sua onestà intellettuale, beh, ne sarò contento.

Vorrei ringraziare anche tutti i miei amici per le risate, per le cose serie ed anche per quelle meno serie. Vorrei sottolineare amici vicini E lontani, anche se con alcuni di loro prendendo strade diverse ci siamo un po’ persi di vista rimangono per me persone con cui ho passato dei bei momenti, e questo mi basta per un ricordo caro ed appassionato. Mi sono sempre sentito rigenerato e con nuova energia dopo una bella serata con voi. Fare la lista sarebbe troppo lungo e sicuramente dimenticherei qualcuno che invece terrei a ricordare. Sono sicuro che chi ne é coinvolto lo sa e conosce anche il perché.

Stefano
Summary

The purpose of my thesis has been to try to fill the gap and contribute to the development of a solid bridge between two research areas which in recent years have been approaching more than ever: quantum optics and semiconductor ultrafast spectroscopy.

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 has been deeply debated topics. Recent proposals [1] and realizations [2, 3, 4, 5, 6] of many-particle entangled quantum states require a 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 the 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
in controlling fabrication processes has enabled the manipulation of the interaction between light and semiconductors by engineering, in addition to the electronic wave functions, the photon states. The radiation rate of an excited atom can be controlled by changing the distribution of electromagnetic modes near the atom by use of 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 immersed 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 modifications of spontaneous emission in a cavity and the strong-coupling regime are cavity quantum electrodynamics effects already well known in quantum optics, these are some of the evidences for the strong similarities existing between semiconductor microcavities and atomic cavity-systems. Entanglement is one of the key features of quantum information and communication technology [10] and a hot topic in quantum optics too. Parametric down-conversion is the most frequently used method to generate highly entangled pairs of photons for quantum-optics applications, such as quantum cryptography and quantum teleportation. Rapid development in the field of quantum information requires monolithic, compact sources of nonclassical photon states enabling efficient coupling into optical fibres and possibly electrical injection. Semiconductor-based sources of entangled photons would therefore be advantageous for practical quantum technologies. The strong light-matter interaction in these systems gives rise to cavity polaritons which are hybrid quasiparticles consisting of a superposition of cavity photons and quantum well excitons [11]. Demonstrations of parametric amplification and parametric emission in SMCs [12, 13, 14], together with the possibility of ultrafast optical manipulation and ease of integration of these microdevices, have increased the interest on
the possible realization of nonclassical cavity-polariton states [15, 16, 17, 18, 19]. In 2005 an experiment probing quantum correlations of (parametrically emitted) cavity polaritons by exploiting quantum complementarity has been proposed and realized [17]. Specifically, it has been shown that polaritons in two distinct idler modes interfere if and only if they share the same signal mode so that which-way information cannot be gathered, according to Bohr’s quantum complementarity principle — a subject deeply discussed in quantum optics —.

The crucial role of many-particle Coulomb correlations in semiconductors marks a profound difference from dilute atomic systems, where the optical response is well described by independent transitions between atomic levels, and the nonlinear dynamics is governed only by saturation effects mainly due to the balance of populations between different levels. In planar SMCs, thanks to their mutual Coulomb interaction, pump polaritons generated by resonant optical pumping may scatter into pairs of polaritons (signal and idler) [20, 12, 21], they are determined by the two customary energy and wave vector conservation conditions $2k_p = k_s + k_i$ and $2E_{k_p} = E_{k_s} + E_{k_i}$ depicting an eight-shaped curve in momentum space. At low pump intensities they are expected to undergo a spontaneous parametric process driven by vacuum-fluctuation, whereas at moderate intensities they display self-stimulation and oscillation [12]. 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 [22, 21, 18, 16, 14]. According to these theoretical calculations, parametric processes are already visible in principle as well as the pump is switched on even at very low excitation intensities. On the contrary, in experiments, apart from an elastic contribution due to Rayleigh scattering, there are present various incoherent phenomena competing to each other in the course of the dynamical evolution.
Elastic Rayleigh scattering, being resonant to the pump frequency, may be greatly reduced by appropriate filtering. Thus, what mainly dominates the light emission at low pump intensities is the photoluminescence (PL) due to the incoherent dynamics of the single scattering events driven by the pump itself. Only once the pumping become sufficient the parametric processes start to reveal themselves and to take over pump-induced PL as well. Indeed, usually, parametric emission and standard pump-induced PL cohabit as shown by experiments at low and intermediate excitation density [14]. Moreover, in order to address quantum coherence properties and entanglement [23] the preferred experimental situations are those of few-particles 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. Thus, a microscopic analysis able to account for parametric emission and pump-induced PL on an equal footing is highly desired in order to make quantitative comparison 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 the unwanted detrimental contributions. Dynamics controlled truncation scheme (DCTS) provides a (widely adopted) starting point for the microscopic theory of the light-matter interaction effects beyond mean-field [24] supplying a consistent and precise way to stop the infinite hierarchy of higher-order correlations which always appears in the microscopic approaches of many-body interacting systems.

In Part I the theoretical tools exploited throughout the thesis are addressed in details providing a fresh insight of what is already present in the literature. Part II will be devoted to present a novel approach based on a DCTS-nonequilibrium quantum Langevin description of the open system in interaction with its surroundings.
we published in Ref. [25]. By means of a sort of microscopic quantum fluctuation-dissipation theorem it enables us to include on an equal footing the microscopic description of the scattering channels competing with the coherent parametric phenomena the optical pump induces. We shall apply our method in order to perform a realistic description of light emission taking into account nonlinear parametric interactions, light quantization, cavity losses and polariton-phonon interaction. We shall present a DCTS-quantum Langevin description of parametric emission including incoherent effects and particular attention will be devoted to the case of single pump feed.

Investigation of the ultrafast dynamics has become in recent years a strategic field both in research and from a technological point of view. Recent developments in ultrafast laser physics and technology now allow to study the very initial interaction processes of non-equilibrium carriers in a semiconductor [26, 27] directly related to the microscopic details of the coupling mechanisms. As a consequence time-resolved laser spectroscopy has become an essential precious tool in modern semiconductor physics. The achievement of very high spatial and temporal resolutions in optical spectroscopies of molecules and solids is among the important experimental advancements of recent years. With near-field scanning optical microscopy (SNOM), the spatial resolution is reduced below the diffraction limit and approaches the scale of quantum confinement [28]. Optical spectroscopy thus becomes a powerful probe of the spatial distribution of quantum states in semiconductor nanostructures [29, 30, 31]. An optical excitation has the ability to generate non-equilibrium carrier distributions and time-resolved spectroscopy provides the best mean of determining the temporal evolution of such distribution functions. Indeed, ultrashort optical pulses may generate coherent superpositions of states and the dynamics of such phase-related quantities can be analyzed. Furthermore when combined with
spatial imaging techniques and/or specific low dimensional structures, ultrafast optical spectroscopy becomes a very powerful tool for investigating a wide variety of phenomena related to relaxation and transport dynamics in semiconductors [27].

In Part III we shall provide a near-field theoretical model for optical spectroscopy. We calculate and introduce the time equation for the most generic quantity to be addressed in order to study the optical response under coherent excitation of a generic QW in the weak-coupling case. By means of the DCTS we will be able to give at any order of the coherent exciting field the system of first order differential equations to be solved numerically in order to gather the output optical field witnessing the nonlinear dynamics inside the heterostructure. The first case-study we shall analyzed will be near-field spectroscopy of QW with interface disorder, i.e. naturally occurring quantum dots.

In Part IV we shall address this crucial problem focusing on a particular very promising source of macroscopic entanglement: parametric down-conversion of photons inside an optical cavity. In the literature two main research lines concerning entanglement can be easily found, one more focused on the group-theoretic properties of the Hilbert space under investigation and the other more interested in which feasible measurement may quantify nonclassical and/or entanglement properties. We shall move our considerations within this second paradigm. A relevant point is whether the conflict between classical elements of reality and quantum mechanics may persist at a macroscopic level [33, 34]. Following Ref. [35], on the one hand we shall quantify the detrimental influence of such environment channels and show how self-stimulation may suppress them efficiently. On the other hand we shall tackle the problem of the macroscopic limit and of the emergence of classical elements of reality within a quantum framework. We shall illustrate a counter-example where the emergence of macroscopic local realism (MLR) may be seen as an intrinsic feature of quantum systems, endogenous in the quantum theory itself (even in the presence
of strong entanglement that is the quintessential of non-classicality). In such a case there is no need at all to rely on environment ingredients (like noise and decoherence). Our results, of course, do not imply that macroscopic entangled systems cannot display violations of local realism [36, 37], but that there is a large class of quantum correlation measurements that cannot be used to show them. This is a great debated topic because, actually, inequalities made up with these correlations are the most used mean for testing and even prove the non-classical, i.e. quantum, features of famous experiments [38, 39]. In addition, also for continuous-variable teleportation some authors [40] are wondering whether a truly quantum description is needed or an explanation in terms of classical phase-space correlation suffices. The results presented indicate that MLR may result from the inability of the observer, practically unavoidable for macroscopic systems, to catch the quantized structure of the system. The emergence of macroscopic local realism in the presence of strong entanglement provides insight into the boundary between classical and quantum worlds. These results, with the care that they have been obtained for a Gaussian system, suggest that, despite the feasible realization of systems with a huge amount of entangled particles, the lack of information gathered by coarse-grained observations may lead to the introduction of elements of local realism even in the presence of strong entanglement and in the absence of decoherence.
Contents

Acknowledgements v

Summary 1

I Quantum optics and optical spectroscopy 1

1 Introduction 3

2 Theoretical tools 6

2.1 Excitons in semiconductors .................................................. 9

2.1.1 The photon interaction ...................................................... 18

2.1.2 Acoustic phonon interaction ................................................ 23

2.2 Dynamics Controlled Truncation Scheme ................................. 26

2.3 Born-Markov approximation ................................................... 32

2.4 Heisenberg-Langevin Approach ............................................... 40

2.4.1 Standard quantum Langevin theory ...................................... 40

2.4.2 Lax contribution .............................................................. 43

II Non-classical properties of light in the strong-coupling 1
### Contents

1. **Regime** 47

2. **Atom optics and nonlinear coherent optical response in semiconductor microcavities** 49
   - 3.1 Incoherent dynamics and parametric processes 49

3. **Coherent optical response beyond mean-field in the 1S exciton sector** 54
   - 4.1 The coupled system 55
   - 4.2 Coherent optical parametric processes for polaritons 74

4. **Incoherent dynamics** 81
   - 5.1 Acoustic phonon interaction 84
   - 5.2 The radiative scattering channel and the coherent pumping 89

5. **Nonequilibrium quantum Langevin approach to parametric emission** 93
   - 6.1 Non-classical properties of microcavity polaritons: analytical study 101
   - 6.2 Photoluminescence dynamics: numerical results 104

6. **Coherent and incoherent optical properties in the weak-coupling case.** 119
   - Application to near-field optical spectroscopy 121

7. **Introduction** 126

8. **Near-field optical spectroscopy: photoluminescence at any exciting order** 126
   - 8.1 Dark states photoluminescence in naturally occurring quantum dots: far- and near-field spectroscopy 130
Part I

Quantum optics and optical spectroscopy
Chapter 1

Introduction

A lot of effort in the past decades has been addressed to atom-cavity systems used to investigate quantum dynamical phenomena in open quantum system and to explore quantum behaviours that have no classical counterparts. The astonishing development of crystal growth techniques has made feasible to investigate even cavity-embedded interacting electron systems by placing quantum wells (QW) in a semiconductor planar microcavity (SMC). This way, Cavity Quantum Electrodynamics (cavity QED) became an interesting and considered research area. The exciton-cavity system arisen 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 if the latter 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 a one-exciton state interacts, in addition owing to customary conservation rules, this exciton has the same wave vector of the impinging beam. In semiconductor heterostructures the nonlinear dynamics does not maintain this simple picture, on the contrary to two-levels atomic systems where the source of nonlinearities comes from saturation, indeed they are mainly due to Coulomb correlation between electrons.
Semiconductors can be structured on a nanometer scale, and thus one can produce materials with tailored properties realizing a wide variety of physically distinct situations. 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. 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 exciting with ultrashort optical pulses in general results in the creation of coherent superpositions of many-particle states. 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 [41, 24, 23, 17]. 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 [42]. The physical picture describing the dynamical evolution of optically excited electrons and holes can be put up in simple terms as follows. First of all, the exciting laser field with frequency near the fundamental band gap creates a 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 give polarization decay and decoherence. Radiative recombination occurs on longer time scales thus the properties of the outgoing light manifest the former electronic dynamics. Ultrafast time resolved spectroscopy has been used as a versatile and
powerful tool in 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 question on light-matter interaction in many-body quantum systems, as well as for appealing future application in optoelectronic and photonic devices.

The mean-field (MF) is a cornerstone of the ultrafast dynamics at the semiconductor band-edge [41]. However, this level of the theory does not provide for a microscopic description of key effects in semiconductor quantum optics as Coulomb scattering and two-pair correlations. The Dynamics Controlled Truncation Scheme (DCTS) provides a (widely adopted) starting point for the microscopic theory of the light-matter interaction effects beyond mean-field [41] supplying a consistent and precise way to stop the infinite hierarchy of higher-order correlations which always appears in the microscopic approaches of many-body interacting systems. In 1996 the DCTS was extended in order to include in the description the quantization of the electromagnetic field [20]. This extension has been applied to the study of quantum optical phenomena in semiconductors as polariton entanglement [19]. However in these works damping has been considered only at a phenomenological level.

In this part we shall analyze in great details the theoretical frameworks and tools we shall use throughout this work.
Chapter 2

Theoretical tools

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 the 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 a bulk semiconductor the conservation of the pseudo-momentum implies that an exciton with wave vector $k$ can interact only with a photon of the same wave vector [43]. On the contrary to atomic systems, a radiative decay is not present and the energy oscillates between the exciton and the photon until, eventually, decaying via non-radiative processes. This coupled propagation modifies their dispersion and the
properties of their optical emission, this is the signature for bulk exciton-polaritons.

Among the various types of nanostructures, quantum wells (QW) allow for the confinement of excitons along only one dimension, they determine a lacking of symmetry in the growth direction (on the following called $z-$axis), thus only $k^\parallel$, the orthogonal component of the wave vector, is conserved. As a consequence an exciton with a given $k^\parallel$ can interact with a light mode on the same $k^\parallel$, but with generic $k_z$ and the radiative decay becomes analogous to the spontaneous emission of an atomic electronic levels. When one or more QWs are immersed in an optical semiconductor microcavity 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. Here Rabi oscillations in the system appear because the excitation oscillates between the exciton and the cavity mode before decaying. The modifications of spontaneous emission in a cavity and the strong-coupling regime are cavity quantum electrodynamics effects already well known in quantum optics, these are some of the evidences for the strong similarities existing between semiconductor microcavities and atomic cavity-systems. However, the crucial role of correlations marks a profound difference from dilute atomic systems, where the optical response is well described by independent transitions between the atomic levels, and the nonlinear dynamics is governed by saturation effects mainly due to level population inversions.

Indeed in semiconductor planar microcavities, thanks to their mutual Coulomb interaction, pump polaritons generated by resonant optical pumping can scatter into pairs of polaritons (signal and idler). At low pump intensities they are expected to undergo a spontaneous parametric process driven by vacuum-fluctuation, whereas at moderate intensities they display self-stimulation [12]. However they are real electronic excitation 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.
2.1 Excitons in semiconductors

Usually, in order to formulate the many-electron problem for solids in the light of providing a description for the excited state in a semiconductor and their coupling with the radiation field, one is led naturally to prefer a second-quantization formalism. The model accounts for electrons as moving in a strictly periodic potential, where the ions are assumed fixed in a Born-Oppenheimer approximation. It means one assumes that the electrons of the inner atomic shells and the positive atomic nuclei give rise to a potential \( V_L \) with the same periodicity of the lattice. Even if the general formalism may be extended in order to include many types of interaction as magnetic ones, in the following we shall retain explicitly only Coulomb interactions between electrons in a semiconductor material.

The Hamiltonian of the system consists of three main contributions which are the kinetic energy of the delocalized electrons, the potential energy in the periodic crystal lattice, and the Coulomb interaction between electrons. In the second quantization form it reads

\[
\hat{H}_c = \int \hat{\psi}^\dagger(\mathbf{r}) \left( -\frac{\hbar^2}{2m} \nabla^2 + V_L(\mathbf{r}) \right) \hat{\psi}(\mathbf{r}) d^3r + \frac{1}{2} \int \int \hat{\psi}^\dagger(\mathbf{r}) \hat{\psi}^\dagger(\mathbf{r}') \frac{e^2}{\varepsilon_r |\mathbf{r} - \mathbf{r}'|} \hat{\psi}(\mathbf{r}') \hat{\psi}(\mathbf{r}) d^3r d^3r',
\]

(2.1)

where \( \hat{\psi}^\dagger(\mathbf{r}) \) and \( \hat{\psi}(\mathbf{r}) \) are the so-called field operators, they obey Fermi commutation rules and they may be thought of as the creation (annihilation) of an electron in the state \( \mathbf{r} \). Still in a very generic case they can be represented on a complete bases set of state \( \{\phi_m(\mathbf{r})\} \):

\[
\begin{align*}
\hat{\psi}^\dagger(\mathbf{r}) & = \sum_m \hat{a}_m^\dagger \phi_m(\mathbf{r}) \\
\hat{\psi}(\mathbf{r}) & = \sum_m \hat{a}_m \phi_m^\ast(\mathbf{r}),
\end{align*}
\]

(2.2)

where \( \hat{a}_m^\dagger \) and \( \hat{a}_m \) creates and destroys an electron in the state \( m \).
A common way to determine these single particle wave functions $\phi_m^*(r), \phi_m(r)$ is a self-consistent Hartree-Fock approximation scheme. Its idea is very easy: once a set of wave functions is given, they are referred to as the zeroth order approximation, this set generates a potential field through the charge distribution which it represents; this potential is used to determine new wave functions, this time, formed within the field of atomic cores and the above charge distribution; again the new set determines a new charge distribution potential and one can go on iteratively. The idea is to start with an uncorrelated many-body electron state and calculate the condition the new set of wave functions must fulfill through the minimization of the expectation functional of the total energy of the system: the uncorrelated state is of course an inaccurate approximation for it neglects the important correlations between electrons, if all the same we wish to obtain the optimum energy (the minimum energy by the variational principle), we should take the unknown wave function $\phi_m(r)$ as our parameters and minimize the total energy with the customary normalization condition on the quantum state [44].

The Hartree-Fock equation obtained this way has the highly evocative form of a Schrödinger equation

$$\left( -\frac{\hbar^2}{2m} \nabla^2 + V_L(r) + \tilde{V}(r) \right) \phi_m(r) - \sum_{m'} A_{m,m'}(r) \phi_{m'}(r) = E \phi_m(r), \quad (2.3)$$

where, as expected,

$$\tilde{V}(r) = \sum_{m'} \int |\phi_{m'}(r)|^2 \frac{e^2}{\varepsilon_r |r - r'|} d^3 r, \quad (2.4)$$

shall be interpreted as the electrostatic potential originating form the charge distribution of the electrons in the $m'$ states, and

$$A_{m,m'} = \int \phi_{m'}(r) \frac{e^2}{\varepsilon_r |r - r'|} \phi^*_m(r) d^3 r \quad (2.5)$$
is the *Coulomb exchange interaction* term due to the fermionic character of the electronic system. The set of equations (2.3) is nonlinear and has to be solved self-consistently with numerical procedures.

For the case of semiconductor structures, the energy spectrum $\epsilon_m$ of the noninteracting carrier Hamiltonian is always characterized by two well-separated energy regions called valence and conduction bands. 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 the 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 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. The Hamiltonian (2.1) describes a two-band direct-gap semiconductor with field operators expanded as

$$
\hat{\psi}^\dagger(\mathbf{r}) = \sum_k \hat{a}^\dagger_{v,k} \phi_{v,k}(\mathbf{r}) + \sum_k \hat{a}^\dagger_{c,k} \phi_{c,k}(\mathbf{r})
$$

$$
\hat{\psi}(\mathbf{r}) = \sum_k \hat{a}_{v,k} \phi^*_v(\mathbf{r}) + \sum_k \hat{a}_{c,k} \phi^*_c(\mathbf{r}), \quad (2.6)
$$

assuming these states to be Hartree-Fock solutions already determined we obtain

$$
\hat{H}_c = \sum_{k,i,j=c,v} E_{i,j,k} \hat{a}_{i,k}^\dagger \hat{a}_{j,k} + \frac{1}{2} \sum_{i,j,l,m=c,v} \delta_{k_i+k_j,k_l+k_m} W_{i,j,k_l,k_m}^{l,m} \hat{a}_{i,k_i}^\dagger \hat{a}_{j,k_j}^\dagger \hat{a}_{l,k_l} \hat{a}_{m,k_m}, \quad (2.7)
$$

with

$$
E_{i,j,k} = \int \phi_{i,k}(\mathbf{r}) \left( -\frac{\hbar^2}{2m} \nabla^2 + V_L(\mathbf{r}) \right) \phi_{j,k}^*(\mathbf{r}) d^3r \quad (2.8)
$$
and
\[ W_i^{jlm} = \int \int \phi_i \phi_j \frac{\epsilon^2}{|r-r'|} \phi^*_m(r) dr dr'. \] (2.9)

Here we are neglecting the terms involving virtual interband transitions which describe the polarization of the electronic orbitals and give mainly contribution to the dielectric constant in the Coulomb potential, it is justified if the excitation binding energy is small compared to the band gap [45, 46]. Along this line and introducing the hole picture (\( \hat{a}_{v,k} = \hat{a}_{h,-k}^\dagger \)) and Hermitian conjugate) we obtain

\[ \hat{H}_c = W_{\text{full}} + \sum_k E_{c,k} \hat{a}_{c,k}^\dagger \hat{a}_{c,k} + \sum_k E_{h,k} \hat{a}_{h,k}^\dagger \hat{a}_{h,k} \]
\[ + \frac{1}{2} \sum_{k',k'',k',k''} W^{cccc}_{k',k'',k',k''} \hat{a}_{c,k'}^\dagger \hat{a}_{c,k'} \hat{a}_{c,k''}^\dagger \hat{a}_{c,k''} + \frac{1}{2} \sum_{k',k'',k',k''} W^{vvcv}_{-k',-k'',-k',-k''} \hat{a}_{h,k'}^\dagger \hat{a}_{h,k'} \hat{a}_{h,k''}^\dagger \hat{a}_{h,k''} - \]
\[ - \sum_{k',k'',k',k'',k} \left( W^{cccc}_{k',-k',-k',-k''} - W^{vvcv}_{k',-k',-k',-k''} \right) \hat{a}_{c,k'}^\dagger \hat{a}_{c,k'} \hat{a}_{h,k} \hat{a}_{c,k'} \] (2.10)

\[
\begin{align*}
W_{\text{full}} &= \sum_k E_{c,k} + \frac{1}{2} \sum_{k',k''} \left( W^{vvcv}_{k',k'',k',k''} - W^{vvcv}_{k',k',k'',k''} \right) \\
E_{h,k} &= -E_{c,k} - \sum_{k',k''} \left( W^{vvcv}_{k',-k',-k'',k'} - W^{vvcv}_{k',-k',-k'',k'} \right) \\
E_{c,k} &= E_{c,0} + \sum_{k',k''} \left( W^{vvcv}_{k',k',k',k''} - W^{vvcv}_{k',k',k',k''} \right)
\end{align*}
\] (2.11)

where the delta of (2.7) is understood but not written explicitly.

For most cases of experimental importance (we will deal with heterostructures) we may assume that electrons and holes lie near the edge of the bands and therefore we may make use of an effective mass approximation and write

\[ E_{c,k} = E_{c,0} + \frac{\hbar^2 k^2}{2m} ; E_{h,k} = -E_{v,0} - \frac{\hbar^2 k^2}{2m} , \] (2.12)

where \( m_e, m_v \) are referred to as electron and hole effective masses.

Usually one is led to consider to take the zero energy reference as the top of the valence band at \( k = 0 \) (which means \( W_{\text{full}} \)) and write
E_{c,k} = E_{\text{gap}} + \frac{\hbar^2 k^2}{2m_c} \quad (2.13)
E_{h,k} = -\frac{\hbar^2 k^2}{2m_v} \quad (2.14)

with \( E_{\text{gap}} = E_{c,0} - E_{v,0} \).

We shall be interested to the optically excited electronic states, in the range of the optical frequencies the wave vector of the photon is negligible with respect to the typical Brillouin zone linear dimension, i.e. \( k_{\text{opt}} \ll (2\pi)/l_0 \) and only exciton with \( k = k_{\text{opt}} \) can be created. As a consequence for delocalized Wannier excitons we can consider only wave vector around zero to be of importance. Due to the symmetry of the crystal lattice the single-particle wave function are Bloch function:

\[ \phi_{n,k}(r) = \frac{1}{\sqrt{V}} e^{i k \cdot r} u_{n,k}(r), \quad (2.15) \]

where \( n \) is the band index and \( u_{n,k}(r) \) is periodic with the same periodicity of the lattice, \( V \) is the quantization volume. Moving along this framework in the integrals (2.9) all the terms involved can be considered as slowly varying in \( r \), all but the periodic part of the Bloch functions. Taking the Taylor expansion of \( W \), retaining only the first term because of the smallness of the \( k \) involved and applying the orthonormality properties of the \( u_{n,k}(r) \), we can rearrange and obtain

\[ W_{\text{ccc}}^{c}\text{c} = W_{\text{c}\text{c}e}^{e}\text{e} = W_{e\text{c}\text{c}}^{e} = W_{e\text{c}\text{e}}^{e} \approx \frac{4\pi e^2}{V \varepsilon r q^2} = V_q, \quad (2.16) \]

the implicit delta function now reads \( q = |k_i - k_j| = |k_i - k_m| \). Finally, we shall set \( W_{k_i,-k_i,-k_m,k_j}^{c}\text{c} \approx 0 \) for it describes the Coulomb exchange interaction and can be shown it tends rapidly to zero for sufficiently small values of \( k \), i.e. for sufficiently distant electron and hole [44].

This is true for bulk semiconductors, but also holds in low-dimensional structures in the infinitely high barriers limit where the confinement wave functions for electron
and holes are the same. However, for finite barriers the matrix elements (2.9) become significantly different for $q$ values larger than the inverse confinement length [46], but nevertheless we shall consider the situation where the small-wavelength behaviour dominates, thus the above is still a good approximation. The value of $\varepsilon_r$ depends on the treatment of the phonon dynamics, for it coincides with the optical dielectric constant whenever phonon-induced correlations due to the Fröhlich interaction are taken into account dynamically, otherwise the static value is the most appropriate choice.

Summarizing, the Hamiltonian for our direct-gap two band semiconductor has the form:

$$\hat{H}_c = \hat{H}_0 + \hat{V}_{\text{Coul}}.$$  \hfill (2.17)

It comprises the single-particle Hamiltonian terms for electron in conduction band and holes in valence band:

$$\hat{H}_0 = \sum_k E_{c,k} \hat{a}_{c,k}^\dagger \hat{a}_{c,k} + \sum_k E_{h,k} \hat{a}_{h,k}^\dagger \hat{a}_{h,k}$$  \hfill (2.18)

and the Coulomb interaction term of three contributions, the two repulsive electron-electron (e-e) and hole-hole (h-h) terms and the attractive (e-h) one:

$$\hat{V}_{\text{Coul}} = \frac{1}{2} \sum_{q \neq 0} \sum_{k,k'} V_q \hat{a}_{c,k+q}^\dagger \hat{a}_{c,k'}^\dagger - \hat{a}_{c,k'} \hat{a}_{c,k} + \frac{1}{2} \sum_{q \neq 0} \sum_{k,k'} V_q \hat{a}_{h,k+q}^\dagger \hat{a}_{h,k'}^\dagger - \hat{a}_{h,k'} \hat{a}_{h,k} - \sum_{q \neq 0} \sum_{k,k'} V_q \hat{a}_{c,k+q}^\dagger \hat{a}_{h,k'}^\dagger - \hat{a}_{h,k'} \hat{a}_{c,k}.$$  \hfill (2.19)

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

$$\hat{H}_c | \Psi \rangle = (\hat{H}_0 + \hat{V}_{\text{Coul}}) | \Psi \rangle = E | \Psi \rangle,$$  \hfill (2.20)
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. $\hat{N}_e = \sum_k \hat{a}_{c,k}^\dagger \hat{a}_{c,k}$ and $\hat{N}_h = \sum_k \hat{a}_{h,k}^\dagger \hat{a}_{h,k}$. Therefore the state $|\Psi\rangle$ may be build up corresponding on a given number of electrons and of holes. Moreover, because we shall consider the case of intrinsic semiconductors materials where $N_e = N_h \equiv N$, the good quantum number for the Schrödinger equation (2.20) is the total number of electron-hole pairs $N$, explicitly

$$\hat{H}_c | N,\alpha \rangle = E_{N,\alpha} | N,\alpha \rangle ,$$

(2.21)

where $\alpha$ is the whole set of proper quantum numbers needed to specify univocally 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:

$$\hat{H}_0 | N,a \rangle = \epsilon_{N,a} | N,a \rangle ,$$

(2.22)

where $N$ identifies the N-pairs subspace and $a$ is a compact form for all the single particle indexes, i.e. $a \equiv k_{e1}, k_{e2}, \ldots, k_{eN}, k_{h1}, k_{h2}, \ldots, k_{hN}$. Indeed

$$| N,a \rangle = \otimes_{n=1}^N \hat{a}_{c,k_{en}}^\dagger \hat{a}_{h,k_{hn}}^\dagger | 0,0 \rangle \text{ and } \epsilon_{N,a} = \sum_{n=1}^N (\epsilon_{k_{en}} + \epsilon_{k_{hn}}) .$$

(2.23)

Being a complete orthonormal basis for the N-pairs subspace we may expand the many-body state $| N,\alpha \rangle$ over it, it yields

$$| N,\alpha \rangle = \sum_a U_a^{Na} | N,a \rangle .$$

(2.24)

It is only a matter of calculation to show that $U_a^{Na}$ is nothing but the enveloping function of the N-pair aggregate, solution of the corresponding Schrödinger equation.
in first quantization. Indeed the eigenvalue problem (2.21) is transformed into:

\[ \sum_{a'} (\langle N,a | \hat{H}_e | Na' \rangle - E_{N,a} \delta_{a,a'} ) U_{a'}^{N} = 0 \]  

(2.25)

In the case of one electron-hole pair, namely the exciton subspace, we could take \( \alpha = (n,\sigma,k) \) and \( a = k_e,k_h \), eq. (2.25) becomes

\[ \sum_{k'_e,k'_h} (\langle k_e,k_h | \hat{H}_e | k'_e,k'_h \rangle - E_{n,\sigma,k} \delta_{k_e,k_h,k'_e,k'_h} ) U_{n,\sigma,k}^{k'_e,k'_h} = 0 \]  

(2.26)

which is the generic Wannier equation for excitons, in the (unrealistic) case of equal effective masses we obtain the result quoted in [47], generalizing to different masses:

\[ | n\sigma k \rangle = \sum_{k'} q^{n\sigma k} \hat{a}_{e,k}^\dagger \hat{a}_{h,-k'+\eta}^\dagger | 0 \rangle \]  

(2.27)

with the ”rules” \( k = k_e + k_h \); \( k' = \eta_e k_e - \eta_h k_h \), \( \eta_{e,h} = m_{e,h}/(m_{e,h} + m_{e,h}) \). Anyway we obtain an even more evocative form if we work directly in the direct lattice \( r \leftrightarrow r_i \) (the former is a continuous variable whereas the latter is a point in the 3D lattice). Using the general mapping [48] \( \sum_{r_i} \leftrightarrow (1/v_0) \int d^3 r \), \( \delta(r-r') = \delta(r_i-r_i)/v_0 \), \( v_0 \) the unit cell volume, it reads

\[ v_0^2 \sum_{r'_e,r'_h} \left( \langle r_e,r_h | \hat{H}_e | r'_e,r'_h \rangle - E_{n,\sigma,k} \frac{\delta_{r_e,r_h,r'_e,r'_h}}{v_0^2} \right) U_{n,\sigma,k}^{r'_e,r'_h} = 0 \]  

(2.28)

with

\[ \langle r_e,r_h | \hat{H}_e | r'_e,r'_h \rangle = \left( -\frac{\hbar^2}{2m_e} \nabla^2_{r_e} - \frac{\hbar^2}{2m_h} \nabla^2_{r_h} - \frac{\epsilon^2}{\varepsilon_r |r_e-r_h|} + V(r_e,r_h) \right) \frac{\delta_{r_e,r_h,r'_e,r'_h}}{v_0^2}, \]  

(2.29)

here \( V(r_e,r_h) \) represents all the additional potential, e.g. those of the heterostructures or those of disorder effects. In the the case we will study deeply in part II of a quantum well without disorder we have \( V(r_e,r_h) = V^e(z_e) + V^h(z_h) \), the potential profile of the quantum well. With the notation \( r = (r^e,z) \), \( R = (m_e r^e + m_h r^h)/(m_e + m_h) \) and \( \rho = (r^e - r^h) \) we obtain
being $M = m_e + m_h$ and $\mu = (m_e + m_h)/m_e m_h$. It gives a factorized exciton wave function

$$U^{n\sigma k}(r_e, r_h) = e^{\frac{i k \cdot R}{\sqrt{A}}} W_{n\sigma}(\rho) \chi_e(z_e) \chi_h(z_h)$$

and

$$| n\sigma, k \rangle = \frac{v_0}{\sqrt{A}} \sum_{r_e, r_h} e^{\frac{i k \cdot R}{\sqrt{A}}} W_{n\sigma}(\rho) \chi_e(z_e) \chi_h(z_h) a^\dagger_{c, r_e} a^\dagger_{h, r_h} | 0 \rangle.$$ 

The most generic representation of a quasi-two-dimensional exciton state with total in-plane center of mass (CM) wavevector $k$ may be set as [49]

$$| \lambda, k \rangle = \sum_{k' k''} e^{\frac{i k \cdot R}{\sqrt{A}}} F_{\lambda}(r_{e, k'}) r_{e, k''} a^\dagger_{c, r_{e, k'}} a^\dagger_{h, r_{h, k''}} | 0 \rangle,$$

$v_0$ and $A$ are the volume of the unit cell and the in-plane quantization surface, whereas $a_{c, r}^\dagger$ ($a_{c, r}$) are creation (annihilation) operator of the conduction- or valence-band electron in the Wannier representation. $r_{e/h} = (r_{e/h} ||, z_{e/h})$ are to be considered coordinates of the direct lattice, $| 0 \rangle$ is the crystal ground state and $R$ the exciton center of mass coordinate $R = (m_e r_e + m_h r_h)/(m_e + m_h)$ with $m_e$ and $m_h$ the effective electron and hole masses. With the usual transformation to Bloch representation we obtain:

$$| \lambda, k \rangle = \sum_{kk'} f_{\lambda}(\mathbf{k} - \mathbf{k'}) \delta_{\mathbf{k}'' - \mathbf{k}', \mathbf{k}} a^\dagger_{e, \mathbf{k}'} a_{e, \mathbf{k}''} | 0 \rangle$$

which is in another form eq. (2.27) provided the transformation to the hole picture ($a_{e, \mathbf{k}} = a_{h, -\mathbf{k}}$ and $-\mathbf{k} |_{el} = \mathbf{k} |_{hole}$).
2.1.1 The photon interaction

Weak coupling

In quantum well, the interaction of the semiconductor with the light field in the usual dipole and rotating wave approximation can be written as

\[ \hat{H}_{\text{int}}^{\text{em}} = -\sum_{\sigma} \int d^3r \hat{E}_\sigma^+(r) \hat{P}_\sigma^-(r) + \text{H.c.} \] (2.35)

We separate the field operator \( \hat{E}(r) \) into a fluctuating part \( \hat{E}_k \) (the one determining the spontaneous emission), that can be expanded in terms of annihilation photon operators as

\[ \hat{E}_k^+ = \alpha_k \hat{a}_k \] (2.36)

and into a classical contribution \( \hat{E}_{\text{sem}}(r) \), describing the (possibly inhomogeneous) exciting electric field. The field operator reads then

\[ \hat{E}^+(r) = \sum_{k\sigma} \hat{E}_k^+ e^{ikr} + \hat{E}_{\text{sem}}^+(r) \] , (2.37)

which in turn means that

\[ \langle \hat{E}_{\text{sem}}^+ \rangle = 0 \] (2.38)

In considering the polarization dependence in exciton terms

\[ P_{\sigma}^+(r) = \sum_{kn} P_{\sigma k n}^+ e^{ikr} = \sum_{k} P_{\sigma k}^+ e^{ikr} = \sum_{k} \frac{1}{\sqrt{V}} \sum_{n} M_{n\sigma}^* B_{n\sigma k} e^{ikr} \] (2.39)

where

\[ M_{n\sigma}^* = \mu_{\sigma}^* \sum_{k'} \Phi_{n\sigma k'} \] (2.40)

we can deal with two Hamiltonians, the quantum and the semiclassical ones:

\[ H_{LM}^{\text{q}} = -V \sum_{k\sigma} E_{\sigma k}^+ P_{\sigma k}^- + \text{H.c.} \]
\[ H_{LM}^{\text{sem}} = -V \sum_{k\sigma} E_{\sigma k}^+ P_{\sigma k}^- + \text{H.c.} \] (2.41)
Usually a classical external field is modeled as \( E_{\sigma,k} = \sum_{\sigma,k} e^{ikr} e^{-i\omega_k t} \epsilon_{\sigma,k} E_{\sigma,k} + c.c. \)

In the dipole and rotating wave approximation is usual to give the classical light-matter interaction Hamiltonian as

\[
\hat{H}_i = -V \sum_{\sigma} \hat{P}_{\sigma}^+ E_{\sigma}^*(t) + H. c.,
\]

where \( E_{\sigma}^*(t) = \sum_k E_{\sigma,k}(t) \). The total Hamiltonian \( \hat{H}_c + \hat{H}_i \) governs the dynamics of the electron system interaction with an external classical light field [24]. However, in applying strictly this way such a dipole approximation to semiconductors, propagation effects inside the medium has been completely neglected. In the case of an extended system as semiconductors really are, in order to apply correctly the dipole approximation one should divide the whole medium into many cells of the same volume, say \( \Omega \) and consider a dipole approximation at each cell [50, 47], provided the cells to be much smaller than the wavelength of the electric field. Then the field-system interaction can be expressed as

\[
\hat{H}_i = -V \sum_{\sigma,k} \hat{P}_{\sigma,k}^+ E_{\sigma,k}^*(t) + H.c.
\]

The multipolar form of the light-matter interaction Hamiltonian for a charged particle systems in interaction with a light field is known as the Power-Zineau-Wolley (PZW) interaction Hamiltonian [48]. This form applied to an electron system interaction with a quantized radiation field is different form the Hamiltonian above even considering a quantum operator instead of a classical light field [48]. Rather surprisingly it contains a field-independent term. Its second quantization form in rotating and cell-dipole approximation then reads

\[
\hat{H}_I = -\frac{V}{\epsilon_0} \sum_{\sigma,k} \hat{D}_{\sigma,k}^+ \hat{P}_{\sigma,k}^- + \frac{V}{2\epsilon_0} \sum_{\sigma,k} \hat{P}_{\sigma,k}^+ \hat{P}_{\sigma,k}^- + H.c.,
\]

where \( \hat{D}_\perp(r) = \sum_{\sigma,k} e^{ikr} \hat{E}_{\sigma,k} \hat{D}_{\sigma,k}^+ + H.c. \) is the so-called transverse displacement field operator [48], proportional to the canonical momentum \( \hat{P} = -\hat{D}_\perp(r)/\epsilon_0 \) conjugate
to the vector potential $\mathbf{A}$ of the light field. It commutes with all the material system operators and can be expressed in terms of photon creation and destruction operators [48]. It is worth underlying that, although the quantum interaction Hamiltonians (2.43) and (2.44) have different structures, the resulting set of equations of motion are compatible with respect to the principle of correspondence and symmetrization [47]. The DCTS as it is addressed in Chapter 2.2 holds for both these Hamiltonian as well. The real-space expression of eq. (2.43) has been used in eq. (2.35).

### QW in semiconductor microcavities

The general theory for calculating polariton modes studying the coupling between excitons and the cavity modes has been presented for the first time by Savona and Tassone [51]. In general, the previous theoretical studies on planar microcavities rely on the assumption that the exciton population which is created with non resonant excitation in a typical experiment can be modeled by an emitting dipole [12, 13, 141] describing the exciton-radiation coupling in perturbation theory, using the Fermi golden rule. Actually, QW excitons obey the crystal momentum conservation rule along the QW plane: an exciton with a given in-plane wavevector $k_\parallel$ interacts only with photon modes having the same $k_\parallel$ and all the possible values of the orthogonal component $k_z$. Provided that the in-plane coherence length of the exciton center-of-mass motion is large, the emission is highly concentrated along the direction determined by the $k_\parallel$ selection rule, and cannot be described by a dipole-like emission pattern. Furthermore, when the exciton-radiation coupling is larger than both the width of the cavity mode and the non-radiative exciton broadening, a strong coupling regime occurs in which the interaction process cannot be described by a simple decay of the exciton level [11, 52]. In this case the Fermi golden rule is not valid and a polariton formalism has to be used instead [17]. In [51] a general quantum mechanical treatment of QW excitons coupled to the electromagnetic
modes of a planar multilayered structure with a given dielectric profile is presented. Firstly, the Maxwell’s equation for the electromagnetic field is solved in the dielectric structure. The resulting eigenmodes are then used to write the field in second quantization form. Then, one QW is placed in an arbitrary position inside one of the dielectric layers and the interaction between the QW exciton and the radiation is studied. The exciton radiation coupling is described by a microscopic hamiltonian where the main exciton features, like the oscillator strength and the in-plane wavevector conservation, are included. This hamiltonian is diagonalized exactly and the dispersion of the resulting polariton modes is obtained. However, it is useful to have some approximation schemes to simply described the light propagation and the resonances in these systems in experimentally achieved situations. It is commonly a good approximation to consider a cavity with symmetric mirrors and equal refractive index inside and outside the cavity. Restricting to the case of normal incidence and for frequencies near the center of the stop band it is possible to parameterize the reflectivity of the Distributed Bragg Reflectors \cite{53} as

\[ r(\omega) = \sqrt{R} e^{i\phi_r(\omega)}, \]  

(2.45)

with \( R \) being the ratio between the reflected and the incident beams, whereas the phase has a linear dependence on the frequency \cite{54}. The polaritonic dispersion is obtain in \cite{51, 54} solving for the poles of an exciton Green function. Expanding close to the resonance one can obtain:

\[ (\hbar \omega - \hbar \omega_{\text{exc}} + i\gamma)(\hbar \omega - \hbar \omega_{\text{c}} + i\gamma_{\text{c}}) = V^2, \]  

(2.46)

where \( \omega_{\text{exc}} \) is the empty-cavity exciton resonance frequency, \( \gamma \) enters in order to take into account the non-radiative exciton broadening, \( \omega_{\text{c}} \) is the cavity mode frequency, \( V \) is known as the vacuum Rabi splitting and \( \gamma_{\text{c}} \) is the halfwidth of the cavity mode.

This equation describes the resonance condition of two coupled and damped harmonic oscillators, the cavity mode and the exciton. It is still valid for cavities
where the light is very confined, on the other hand, because the reflectivity of the DBRs decreases for small incident angles and a stop-band is no more present, it is no more valid for great wavevectors [54], in these cases one has to consider the solutions of the exact dispersion relation and leaky modes arise. Nevertheless, for the data presented in the literature and hence for the usually accomplished experimental works, the two-coupled oscillators equation is a good approximation. Following this line of argument we shall model the interaction of the electron system with cavity modes in the usual rotating wave approximation by

\[ \hat{H}_I = - \sum_{n,k} V_{n,k} \hat{a}_k^\dagger \hat{B}_{n,k} + H.c. \]  

(2.47)

with the operator \( \hat{a}_k^\dagger \) which creates a photon state with \( k = (\sigma, \mathbf{k}) \) and energy \( \hbar \omega_k = \hbar (\omega_{\text{exc}}^2 + v^2|k|^2)^{1/2} \), \( v \) being the velocity of light inside the cavity, \( \hat{B}_{n,k}^\dagger \) creates an exciton state with the same wave vector and polarization \( k \) and energy \( \hbar \omega_{1,n,k} \). \( V_{n,k} \) is the photon-exciton coupling coefficient [54] enhanced by the presence of the cavity.
2.1.2 Acoustic phonon interaction

In the case of GaAs-AlAs QW structures, the electron and hole of the excitons are considered to be well confined within a QW since the bandgap discontinuity is quite large. On the other hand, the lattice properties of such materials are in close proximity, thus the acoustic-phonons which interact with the quasi-two-dimensional exciton can be considered to have three-dimensional character. The three-dimensional (DF) electron-phonon interaction Hamiltonian, can be written as

$$\hat{H}_{\text{e-ph}}^{\text{DF}} = \sum_{\mathbf{k}, \mathbf{q}} \left( \frac{\hbar |\mathbf{q}|}{2 \rho u V} \right)^{1/2} \left( D_c \hat{a}_{\mathbf{c}, \mathbf{k}+\mathbf{q}} \hat{a}_{\mathbf{c}, \mathbf{k}} + D_v \hat{a}_{\mathbf{v}, \mathbf{k}+\mathbf{q}} \hat{a}_{\mathbf{v}, \mathbf{k}} \right) \left( \hat{b}_{\mathbf{q}} + \hat{b}_{-\mathbf{q}}^\dagger \right).$$

(2.48)

Here $\hat{a}_{\mathbf{c}/\mathbf{v}, \mathbf{k}}$, $\hat{a}_{\mathbf{c}/\mathbf{v}}$ are creation and destruction operator of the conduction- valence-band electron in Bloch representation. Our interest is to project such Hamiltonian in exciton bases. A quasi-two-dimensional exciton state with total wave vector $\mathbf{k}^\parallel$ may be represented as

$$|\lambda, \mathbf{k}\rangle = \frac{v_0}{L} \sum_{\mathbf{r}_e, \mathbf{r}_h} e^{i \mathbf{k} \cdot \mathbf{R}} F_\lambda (\mathbf{r}_e^\parallel, \mathbf{r}_h^\parallel, \mathbf{r}_e^\perp, \mathbf{r}_h^\perp) \hat{a}_{\mathbf{c}, \mathbf{r}_e} \hat{a}_{\mathbf{v}, \mathbf{r}_h} |0\rangle,$$

(2.49)

$v_0$ and $L$ are the volume of the unit cell and the linear dimension of the quantization volume, whereas $\hat{a}_{\mathbf{c}/\mathbf{v}, \mathbf{r}}^\dagger (\hat{a}_{\mathbf{c}/\mathbf{v}, \mathbf{r}})$ are creation (annihilation) operator of the conduction- or valence-band electron in the Wannier representation. $\mathbf{r}_e$ and $\mathbf{r}_h$ are to be considered coordinates of the direct lattice, $|0\rangle$ is the crystal ground state and $\mathbf{R}$ the exciton center of mass coordinate

$$\mathbf{R} = (m_e \mathbf{r}_e^\parallel + m_h \mathbf{r}_h^\parallel)/(m_e + m_h),$$

(2.50)

with $m_e$ and $m_h$ the effective electron and hole masses. Transforming into the Bloch representation by means of the relation ($N = v_0 L^3$ is the number of unit cells)
one obtains

$$| \lambda, k \rangle = \sum_{\mathbf{k}, \mathbf{k}' \parallel} f_\lambda(\mathbf{k}, \mathbf{k}'; \mathbf{k}) \hat{a}_{\mathbf{k}||\mathbf{k}'} \hat{a}_\mathbf{k}^\dagger \hat{a}_\mathbf{k} | 0 \rangle ,$$

(2.52)

where the lines are used to differentiate the relative wave vectors of the two constituents from the center of mass wave vector (k).

Projecting, the Hamiltonian describing exciton-acoustic phonon coupling via deformation potential of eq.(2.48) may be set as

$$\hat{H}_{\text{DF}}^{\text{exc}} - \text{ph} = \sum_{q, \parallel} t^q_{\lambda, \mathbf{k}} | \lambda, \mathbf{k} \rangle \langle \lambda, \mathbf{k} + q|| | (b_q + b^\dagger_{-q}) .$$

(2.53)

It is worth saying this projection may be accomplished within any number of electron-hole pairs as it will be used in part III. As we shall analyze in detail in the following, let us consider a usual semiconductor model Hamiltonian $\hat{H}_{\text{sem}} = \hat{H}_0 + \hat{V}_{\text{Coul}}$, composed of a free-particle part $\hat{H}_0$ and the Coulomb interaction $\hat{V}_{\text{Coul}}$. The eigenstates $|N\alpha k\rangle$ with energy $\omega_{N\alpha k}$ of $\hat{H}_c$ can be labeled according to the number of e-h pairs and the total momentum $\mathbf{k}$ [55, 24], this way we shall write it as

$$\hat{H}_{\text{sem}} = \hat{H}_{\text{exc}} = \sum_{N\alpha k} \omega_{N\alpha k} | E_{N\alpha k} \rangle \langle E_{N\alpha k} | .$$

(2.54)

Then, the acoustic phonon interaction Hamiltonian reads:

$$\sum_{Nk\zeta \eta q^||, q_z} t^q_N_{k\zeta \eta} X_{N\zeta (k+q^||); N\eta k} (b_{q^||, q_z} + b^\dagger_{-q^||, q_z}) ,$$

(2.55)

where $\zeta, \eta$ are the proper quantum number spanning the associated N-pair state space (e.g.if $N = 1 \zeta = (n_\zeta, \sigma_\zeta)$). This kind of interaction process conserves the number of electron-hole pairs and thus does not alter the $N$ subspace direct sum
structure of the global Hilbert space. In the Hamiltonian above we have include all the other ingredients inside $t$:

$$
t^q_{Nk;\zeta\eta} = \sum_k \left( \frac{\hbar|q|}{2\rho v_s V} \right)^{1/2} \left( \langle N\zeta(k + q^\|) | D_v \hat{c}^\dagger_{k+q} \hat{c}_k + D_v \hat{d}^\dagger_{k+q} \hat{d}_k | N\eta k \rangle \right) \quad (2.56)
$$
2.2 Dynamics Controlled Truncation Scheme

In any theory aiming at describing the dynamics of semiconductor electrons interacting with a light field one is lead to face an infinite hierarchy of dynamical variables and hence to consider an appropriate truncation procedure.

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 [56]. The standard model Hamiltonian for the optically induced dynamics near the semiconductor band-edge can be written as

\[ \hat{H} = \hat{H}_{\text{semiconductor}} + \hat{H}_{\text{carrier-field}}, \]

where \( \hat{H}_{\text{semiconductor}} \) describes the semiconductor material (coupled to the environment, e.g. lattice vibrations) and \( \hat{H}_{\text{carrier-field}} \) accounts for the interaction of the carriers with electric fields applied to the sample.

The mentioned classification relies on a special property of the Hamiltonian in equation (2.57), namely that the number of pair excitations in the system can change only due to the action of the applied field. Apart from semiconductors, Frenkel exciton systems also (e.g. molecular aggregates, molecular crystals or biological antenna systems) are commonly described in terms of models sharing this property. In fact, DCTS type approaches have been formulated for these systems too [41].

DCTS can be regarded as a systematic way of selecting a set of relevant dynamical variables, once a given order \( p_M \) 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, gives 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 has been clear it has far reaching implementations when combined with other techniques, e.g. projection techniques as those in [24, 20, 47]. Moreover, it can be combined with every strategy to deal with the phonon-assisted branch of the hierarchy [57, 58], 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 theorem and its proof that follow are taken from [47, 20]. It represents the generalization to the full quantum case of a similar procedure [60, 59, 24] made in the semiclassical case.

Let us consider a usual semiconductor model Hamiltonian $\hat{H}_c = \hat{H}_0 + \hat{V}_{\text{Coul}}$, composed of a free-particle part $\hat{H}_0$ and the Coulomb interaction $\hat{V}_{\text{Coul}}$. The eigenstates $|N_\alpha k\rangle$ with energy $\omega_{N_\alpha k}$ of $\hat{H}_c$ can be labeled according to the number of e-h pairs and the total momentum $k$ [55, 24]. The ground state of the semiconductor is defined as the vacuum state with respect to the exciton annihilation

$$
\hat{B}_{nk} |0\rangle = 0, \quad \hat{B}_{nk} = \sum_{k'} \Phi^*_{n\sigma k'} \hat{a}_{h,-k'+\eta_k} \hat{a}_{c,k+k'+\eta_k}
$$

and correspond to the full valence band. Here $\Phi$ is the exciton wave-function and $\hat{a}_c$ and $\hat{a}_h$ are electron and hole annihilation operators. The one-exciton subspace is the closure of the linear span of $|E_n^{(1)}\rangle$ with energy $\omega_{1,n,k}$, it is worth noticing that this way both bound and scattering states are included in $n$, meanwhile $k = (\sigma, k)$. The polarization index $\sigma$ stands to remember which specific optical transition is
required in order to excite the eh-pair and hence to create the exciton, if optically active this correspond to the helicity of the light involved, $k$ is the center of mass wavevector. The next relevant subspace is the biexciton one corresponding to all the 4-particles aggregates. It is built up from the complete basis $|E_m^{(2)}\rangle$ with $w_{2,m}$ where $m$ represents the suitable collection of quantum numbers needed to specify the bound as well the unbound states. The dielectric screening is approximately accounted for by the static dielectric constant of the semiconductor. The eigenstates of the Hamiltonian $\hat{H}_{ph} = \sum_{\sigma k} \hbar \omega_{\sigma k} \hat{a}_{\sigma k}^\dagger \hat{a}_{\sigma k}$ are written as $|n,\lambda\rangle$ where $n$ stands for the total number of photons in the state and $\lambda = (k_1,\sigma_1;...,k_n,\sigma_n)$ specifies wave vector and polarization $\sigma$ of each photon.

The interaction Hamiltonian $\hat{H}_I$ has a different form with respect to the particular system under investigation, i.e. cavity exciton-polariton [20] or Hyper-Raman scattering in semiconductors [47]. Anyway, for the proof of the theorem its explicit form is not needed apart form the fact that the coupling is of linear type

$$\hat{H}_I \propto \hat{a}_r^\dagger \hat{B}_r + H.c.$$ (2.59)

In order to analyze the dynamical evolution of such coupled system we shall consider the expectation values of the following generalized projection operators, also known as Hubbard operators:

$$\hat{X}_{N,\alpha;M,\beta} = |E_{N,\alpha}\rangle \langle E_{M,\beta}|.$$ (2.60)

The theorem states that the expectation values of these operators can be expressed as a power series in the input field ($n = \sum_k n_k$).

$$\langle \hat{X}_{N,\alpha;M,\beta} \prod_{k,k'} \hat{a}_{k}^{n_k} \hat{a}_{k'}^{n_{k'}} \rangle = \sum_{i=0}^{\infty} \langle \hat{X}_{N,\alpha;M,\beta} \prod_{k,k'} \hat{a}_{k}^{n_k} \hat{a}_{k'}^{n_{k'}} \rangle (N+M+n+n'+2i)$$

$$+ \mathcal{O}(\varepsilon^{(N+M+n+n'+2i+2)}) .$$ (2.61)

There are three main reasons for this theorem to hold. The crystal in the absence of an optical excitation can be described by a zero particle system because the state
of the system is $| E_{N=0} \rangle$, thus all the many body correlations are induced by the optical excitation, anyway this condition can be relaxed for it is sufficient to assume the system to be in thermal equilibrium, if in the absence of input [58]. Moreover, the nonlinear and quantum correlation effects among photons are induced by the electron systems and the free Hamiltonians $\hat{H}_S$ and $\hat{H}_{ph}$ preserve the number of electron-hole pairs and the number of photons respectively, i.e. the transitions are due only to $\hat{H}_I$ preserving the total number of quasi-particle (e-h pairs plus photons). If explicitly included in the model, the only term which produces quasi-particles is $\hat{H}_p$ which represents the pump seed to the cavity system generating cavity photons by the perturbative parameter, i.e. the coherent input light beam $\mathcal{E}$. However, for the proof of the theorem the only thing that is needed is to set properly the initial condition to be a coherent state of the radiation.

At $t = t_0$, the state describing the quantized light field contains only photons at the input mode and the electronic system is in its ground state. In an Heisenberg description adopted here as the time goes on the state remains unchanged and the evolution of the coupled electron-photon system is described by Heisenberg equation of motion. For simplicity we shall consider a single mode coherent input field, $\langle \hat{a}_{kp}(t_0) \rangle = \mathcal{E}$. It implies that the initial condition of the system of differential equations must be set as:

$$\langle \hat{X}_{N,\alpha;M,\beta} \prod_{k,k'} \hat{a}_{k}^{\dagger n_k} \hat{a}_{k'}^{n_{k'}} \rangle_{t_0} = \delta_{N+M} \prod_{k,k'} \delta_{(n_k+n_{k'})} \delta_{\mathcal{E}^{n_k n_{k'}} \mathcal{E}^{n_{k'} n_{k}}} \quad (2.62)$$

In the absence of input we are manipulating the zeroth order, for what chosen before we have:

$$\langle \hat{X}_{0;0} \rangle^{(0)} = 1 \leftrightarrow \langle 0 | 0 \rangle^{(0)} = \langle \mathbb{I} \rangle^{(0)}$$

$$\langle \hat{X}_{N,\alpha;M,\beta} \prod_{k,k'} \hat{a}_{k}^{\dagger n_k} \hat{a}_{k'}^{n_{k'}} \rangle^{(0)} = 0 \quad \forall \ N + M + n + n' \neq 0 \quad (2.63)$$
The theorem can be easily proved simply computing the Heisenberg equation of motion for the generic dynamical variable and considering the terms involved in the light of the expansion parameter and of the initial condition. Following [47] we set
\[ \hat{X}_{N,\alpha;M,\beta} \prod_{k,k'} \hat{a}_{k}^{\dagger} \hat{a}_{k'}^{\dagger} = \hat{Z}_{M,n'}^{N,n}. \] Subjected to the Hamiltonian \( \hat{H}_{c} + \hat{H}_{ph} + \hat{H}_{I} \) the time evolution reads:
\[
\frac{d}{dt} \hat{Z}_{M,n'}^{N,n} = c_{M,n'}^{N,n} \hat{Z}_{M,n'}^{N,n} + c_{M-1,n'+1}^{N,n} \hat{Z}_{M-1,n'+1}^{N,n} + \\
c_{M,n'+1}^{N+1,n} \hat{Z}_{M,n'+1}^{N+1,n} + c_{M,n'}^{N+1,n-1} \hat{Z}_{M,n'}^{N+1,n-1} + \\
c_{M+1,n'-1}^{N,n} \hat{Z}_{M+1,n'-1}^{N,n} + c_{M+1,n'}^{N,n+1} \hat{Z}_{M+1,n'}^{N,n+1} + \\
c_{M,n'}^{N-1,n+1} \hat{Z}_{M+1,n'}^{N-1,n}. \quad (2.64)
\]
All the terms but two on the right-hand side belong to the subspace \( p = N + M + n + n' \) to which also \( \hat{Z}_{M,n'}^{N,n} \) belongs. There are two terms, the third and the sixth, belonging to \( p = p + 2 \), thus we are facing an open hierarchy of equations of the type \( p = p + 2i_{0} \) with \( i_{0} \geq 0 \) any generic integer. The most important point of the whole discussion is the following: the set of equations does constitute a homogeneous linear system of first-order differential equations whose solution can be set as a summation of terms depending linearly on the initial conditions. In this case the initial conditions are (2.62) considered at the order \( p = p + 2i_{0} \). The proof is finished just taking the expectation value of this solution combined with (2.62): regarding a dynamical variable of order, say, \( s \), the solution depends on terms of the same order (because of the initial conditions of order \( s \)) and on terms of order \( s + 2 \). The latter depend on terms of their same order \( s + 2 \) (because of the initial conditions of order \( s + 2 \)) plus others of order \( s + 4 \) and so on. Picking again \( i_{0} \) as an arbitrary integer greater or equal to zero we can rearrange the solution dependency of the generic dynamical variables of order \( s \) as
\[
\langle \hat{Z}_{[s]} \rangle = \sum_{i=0}^{i_{0}} \langle \hat{Z}_{[s]} \rangle^{(s+2i)}, \quad (2.65)
\]
where simply $\langle \hat{Z}_{[s]} \rangle^{(s+2i)}$ means the collection of all the contributions to $\langle \hat{Z}_{[s]} \rangle$ of order $(s + 2i)$. For this construction what is left apart are terms of order $(s + 2i_0 + 2)$ in the applied coherent field which is exactly what stated in Chapter 4.2. Once a perturbative order $p_M$ is chosen, the theorem establishes that the hierarchy of expectation values $\langle \hat{Z}_{M,n}^{N,n} \rangle^{(p_M)}$ comprised all (and only) the equations concerning the operators with $\bar{p} \leq p \leq p_M$. 
2.3 Born-Markov approximation

In any microscopic treatment of any solid-state system, we are facing a huge number of degrees of freedom and a statistical description of the problem becomes imperative [61]. The time evolution of a generic physical quantity $O$ – described by the operators $\hat{O}$ – is obtained by the generic rule

$$O(t) = Tr\{\hat{O} \hat{\rho}\},$$

(2.66)

representing a quantum plus statistical average. $\hat{\rho}$ is the so-called density matrix operator and it is defined as the statistical average of the projection operator corresponding to the generic state $|\Psi\rangle$ of the whole system, i.e. $\hat{\rho} = |\Psi\rangle\langle\Psi|$. In the Heisenberg description of the dynamics $\hat{\rho}$ represents a fixed reference frame and are the operators, e.g $\hat{O}$, to have a proper time evolution which, under the trace, gives the time dependence of the observable $O(t)$. Anyway it is only a matter of choice dictated by practical convenience, indeed in the Schrödinger picture, on the other hand, is the reference frame of the states vectors to evolve in time and all the operators do not, whereas the so-called interaction picture sets its steps in the middle of the two. It is very easy to move from one to another by means of a proper unitary transformation [62].

If we want to calculate the time evolution of a generic operator, the trace is an invariant quantity under unitary transformation, thus

$$\frac{d}{dt}O(t) = tr\{\hat{O} \frac{d}{dt}\hat{\rho}(t)\} = tr\{\frac{d}{dt}\hat{O}^H \hat{\rho}^H(t_0)\}.$$  

(2.67)

In order to set up the perturbative expansion is customary to focused on the time evolution of the density matrix. We shall pursue this line of argument believing the physical motivations of certain crucial approximations to become self-evident. We consider a generic physical system weakly interacting with a surrounding. The
The corresponding Hamiltonian can be schematically written as,
\[
\hat{H} = \hat{H}_0 + \hat{H}_{\text{int}},
\] (2.68)
the first term represents the unperturbed system + reservoir Hamiltonian \( \hat{H}_0^S + \hat{R} \) while \( \hat{H}_{\text{int}} \) is the sum of all the environment interaction providing to the system dissipation and dephasing. When an operator is only a system operator, i.e. \( \hat{M} \equiv \hat{M} \otimes 1_R \),
\[
\text{tr}\{\hat{M} \otimes 1_R \hat{\rho}\} = \text{tr}_S\{\hat{M} \hat{\rho}^S\}, \quad \rho^S = \text{tr}_R\{\hat{\rho}\}
\] (2.69)
is the partial trace of the global density matrix over the reservoir degrees of freedom, i.e. the system density matrix operator.

In the Schrödinger picture the density matrix operator time evolution is governed by the Liouville-Von Neumann equation of motion:
\[
\frac{d}{dt}\hat{\rho} = \mathcal{L}(\hat{\rho}) = \frac{1}{i\hbar}[\hat{H},\hat{\rho}] \equiv \left(\hat{H},\hat{\rho}\right).
\] (2.70)
The Hamiltonian separation eq. (2.68) gives
\[
\frac{d}{dt}\hat{\rho} = \frac{d}{dt}\hat{\rho}\bigg|_{\hat{H}_0} + \frac{d}{dt}\hat{\rho}\bigg|_{\hat{H}_{\text{int}}},
\] (2.71)
As soon as one is able to solve exactly the dynamics dictated by \( \hat{H}_0 \) we can pass to the interaction picture where
\[
\hat{\rho}^i(t) = \hat{U}_0^\dagger(t-t_0)\hat{\rho}\hat{U}_0(t-t_0),
\] (2.72)
\[
\frac{d}{dt}\hat{\rho}^i(t) = \left(\hat{H}_{\text{int}},\hat{\rho}^i(t)\right),
\] (2.73)
here \( \hat{U}_0(t-t_0) = \exp(\hat{H}_0(t-t_0)/i\hbar) \) is the unitary evolution operator corresponding to the noninteracting Hamiltonian and
\[
\hat{H}_{\text{int}} = \hat{U}_0^\dagger(t-t_0)\hat{H}_{\text{int}}\hat{U}_0(t-t_0).
\] (2.74)
The key point under any perturbative approach is that the effect of the interaction Hamiltonian \( \hat{H}_{\text{int}} \) is small compared to the free evolution dictated by \( \hat{H}_0 \), a precise notion can be given in interaction picture terms in saying that the interaction matrix elements of \( \hat{H}_{\text{int}}^i \) over the noninteracting bases states of the system+resevoir are smaller than the typical energy difference between the 2 bases states the element connects. Actually, we shall consider a situation where the resevoir correlation time \( \tau_c \) is short compared to all the system relaxation times, i.e. let’s say \( \Gamma^{-1} \), but not zero. On the contrary, in order to have an essentially Markovian behaviour, \( \tau_c \) must be long compared to the reciprocal of the natural frequencies (\( \omega_S^{-1} \)) of the system — the latter may evolve rapidly following its unperturbed motion — and changes are to be observed over time intervals \( \Delta t \) such that

\[
\omega_S^{-1} < \tau_c < \Delta t \ll \Gamma^{-1}.
\]

(2.75)

In this spirit we formally integrate eq (2.73) from a time \( t_0 \) to the current time \( t \)

\[
\dot{\hat{\rho}}^i(t) = \dot{\hat{\rho}}^i(t_0) - \int_{t_0}^{t} dt' \left( \hat{H}_{\text{int}}^i(t'), \dot{\hat{\rho}}^i(t') \right),
\]

(2.76)

where up to now no approximation are made, we shall continue and obtain the well-known Neumann series:

\[
\dot{\hat{\rho}}^i(t) = \dot{\hat{\rho}}^i(t_0) + \sum_{n=1}^{\infty} \int_{t_0}^{t} dt_1 \int_{t_0}^{t_1} dt_2 \cdots \int_{t_0}^{t_{n-1}} dt_n \left( \hat{H}_{\text{int}}^i(t_1), \cdots, \hat{H}_{\text{int}}^i(t_n), \dot{\hat{\rho}}^i(t_0) \right) \right).
\]

(2.77)

For our purpose we will be interested up to the second order. In the spirit of the perturbative approach we are performing the time variation of \( \dot{\hat{\rho}}^i \) can be considered adiabatically slow compared to that of the Hamiltonian \( \hat{H}_{\text{int}}^i \) within the interaction picture. As a consequence, the former can be taken out of the time integral in \( t' \)
and evaluated at the current time $t$. Such an adiabatic limit provides us with the following effective Liouville-Von Neumann equation

$$
\frac{d}{dt} \hat{\rho}_i(t) = \left( \hat{H}_0^i(t), \hat{\rho}_i(t) \right) + \left( \hat{H}_{int}^i(t), \left( \hat{K}_{int}^i(t), \hat{\rho}_i(t) \right) \right), \tag{2.78}
$$

where

$$
\hat{K}_{int}^i(t) = \int_{t_0}^{t} \hat{H}_{int}^i(t')dt'. \tag{2.79}
$$

In the Schrödinger picture it reads

$$
\frac{d}{dt} \hat{\rho} = \tilde{L}(\hat{\rho}) + \hat{C}. \tag{2.80}
$$

$$
\hat{C} = \left( \hat{H}_{int}, \hat{U}_0(t-t_0)\hat{\rho}(t_0)\hat{U}_0^\dagger(t-t_0) \right) \tag{2.81}
$$

represents how the quantum correlation presented at time $t_0$ propagates to $t$, but anyway for the coupling we shall consider it vanishes [61] and

$$
\tilde{L}(\hat{\rho}) = \left( \hat{H}_0, \hat{\rho}(t) \right) + \left( \hat{H}_{int}, \left( \hat{K}, \hat{\rho}(t) \right) \right), \tag{2.82}
$$

where

$$
\hat{K} = \int_{t_0}^{t} \hat{U}_0(t-t')\hat{H}_{int}\hat{U}_0^\dagger(t-t')dt'. \tag{2.83}
$$

In the spirit of a system in interaction with its surrounding we are interested in

$$
\frac{d\hat{\rho}^S}{dt} = tr_R \{ \tilde{L}(\hat{\rho}) \}. \tag{2.84}
$$

In general the trace over the reservoir degrees of freedom does not commute with the Liouville operator $\tilde{L}(\hat{\rho})$ and we cannot build a closed equation of motion for $\hat{\rho}^S$ without other assumption. Typically one considers, at this point, the reservoir subsystem as characterized by a huge number of degrees of freedom with respect to the system $S$ of interest. We shall consider the reservoir $R$ as very much bigger than $S$ so that not to be so significantly perturbed by $S$, i.e. it behaves as a proper
reservoir always in thermal equilibrium. This way the density matrix operator can be set as

\[ \hat{\rho} = \hat{\rho}^S \otimes \hat{\rho}^R, \quad \hat{\rho}^R = e^{-\frac{\hat{R}}{k_B T}} \text{tr}_R \left\{ e^{-\frac{\hat{R}}{k_B T}} \right\}. \] (2.85)

The new effective Liouville operator reads

\[ \hat{\mathcal{L}}^S(\hat{\rho}^S) = \left( \hat{\mathcal{H}}^S_0, \hat{\rho}^S \right) + \text{tr}_R \left\{ \left( \hat{\mathcal{H}}_{\text{int}}, \left( \hat{\mathcal{K}}_i \hat{\rho}^S \otimes \hat{\rho}^R \right) \right) \right\}. \] (2.86)

The typical interaction Hamiltonian we will consider is of the form

\[ \hat{\mathcal{H}}_{\text{int}} = \sum_i \left( \hat{\mathcal{Q}}_i \hat{\mathcal{F}}_i + \hat{\mathcal{Q}}_i^\dagger \hat{\mathcal{F}}_i^\dagger \right). \] (2.87)

with the bath operators of Boson statistics, i.e. \[ [\hat{F}_i, \hat{F}_j] = g_i \delta_{i,j} \] and \[ \langle \hat{F}_i^\dagger \hat{F}_j \rangle = n_R i \delta_{i,j}, \]

\[ i \] is the quasi-particle quantum number parameterizing the system operator. The last term in (2.86) becomes

\[ -\frac{1}{\hbar^2} \sum_i \left( (n_R^i + g_i) \left[ \hat{Q}_i \hat{K}_i^{(+) \dagger} \hat{\rho}^S \right] + n_R^i \left[ \hat{\rho}^S \hat{K}_i^{(+) \dagger} \hat{Q}_i \right] + n_R^i \left[ \hat{Q}_i \hat{K}_i^{(-)} \hat{\rho}^S \right] + (n_R^i + g_i) \left[ \hat{\rho}^S \hat{K}_i^{(-) \dagger} \hat{Q}_i \right] \right), \] (2.88)

where

\[ \hat{K}_i^{(+) \dagger} = \int_0^{t-t_0} \frac{d\tau e^{-\frac{R^\tau}{i \hbar}}}{i \hbar} \hat{Q}_i^\dagger e^{-\frac{R^\tau}{i \hbar}} e^{\frac{R^\tau}{i \hbar}}, \]

\[ \hat{K}_i^{(-) \dagger} = \int_0^{t-t_0} \frac{d\tau e^{-\frac{R^\tau}{i \hbar}}}{i \hbar} \hat{Q}_i e^{-\frac{R^\tau}{i \hbar}} e^{-\frac{R^\tau}{i \hbar}}. \] (2.89)

It is common that the system operator has the general property \[ \hat{K}_i^{(+) \dagger} = \hat{\mathcal{Q}}_i \] [61], in this case we can rearrange in a more compact and evocative form. In (2.89) we can manipulate

\[ \hat{K}_i^{(-) \dagger} = \int_0^{t-t_0} d\tau e^{-\frac{R^\tau}{i \hbar}} \hat{Q}_i e^{-\frac{R^\tau}{i \hbar}} e^{-\frac{R^\tau}{i \hbar}} = \int_0^{t-t_0} d\tau e^{-\frac{R^\tau}{i \hbar}} \hat{Q}_i^\dagger e^{-\frac{R^\tau}{i \hbar}} e^{-\frac{R^\tau}{i \hbar}} = \hat{\mathcal{D}}_i^{(-) \dagger}. \] (2.90)
The last two terms in (2.88) can be manipulated, transforming \( i \rightarrow -i \) and summing in the reverse order \( (n_{R}^{i} = n_{i}^{R} \) without loss of generality) we obtain

\[
- \frac{1}{\hbar^2} \sum_{i} \left( (n_{R}^{i} + g_{i}) \left[ \hat{Q}_{i}, \hat{D}_{i}^{(+)} \hat{\rho}^{S} \right] + n_{i}^{R} \left[ \hat{\rho}_{S}^{+} \hat{D}_{i}^{(-)} \right] \right) +
\]

\[
+ n_{i}^{R} \left[ \hat{Q}_{i}, \hat{D}_{i}^{(-)} \hat{\rho}^{S} \right] + (n_{R}^{i} + g_{i}) \left[ \hat{\rho}_{S}^{+} \hat{D}_{i}^{(+)} \right] =
\]

\[
= - \frac{1}{\hbar^2} \sum_{i, \pm} \left[ \hat{Q}_{i}, \hat{D}_{i}^{(\pm)} \hat{\rho}^{S} - \hat{\rho}_{S}^{\pm} \hat{D}_{i}^{(\mp)} \right] (n_{R}^{i} + g_{i} \theta(\pm)) ,
\]

(2.91)

where, we have defined

\[
\hat{D}_{i}^{(\pm)} = \int_{t}^{t_{0}} dt' e^{\frac{i n_{R}^{i}}{\hbar} Q_{i}^{\dagger}} e^{-\frac{i n_{R}^{i}}{\hbar} Q_{i}^{\dagger}} e^{\pm \frac{i n_{R}^{i}}{\hbar} t'}. 
\]

(2.92)

This term provides, through the complete collision limit, scattering and renormalization effects [61]. As mentioned at the beginning, our aim is to build up Markov approximation in the Heisenberg picture, but the adiabatical decoupling between free carriers and many-body environment interaction is not harmless. Indeed, in order to accomplish this goal, we need to make some steps backwards, just before the adiabatic limit. By inserting eq. (2.76) into eq. (2.73) we obtain an integro-differential equation representing the interaction dynamics with no approximations.

In the Schrödinger picture is

\[
\frac{d}{dt} \hat{\rho}(t) = \left( \hat{H}_{0}, \hat{\rho}(t) \right) + \hat{C} + \int_{t_{0}}^{t} dt' \left[ \hat{H}_{int}, \right.
\]

\[
\left( \hat{U}_{0}(t - t'), \hat{H}_{int} \hat{U}_{0}^{\dagger}(t - t'), \hat{U}_{0}(t - t') \hat{\rho}(t') \hat{U}_{0}^{\dagger}(t - t') \right) ,
\]

(2.93)

considering again \( \hat{\rho}(t') = \hat{\rho}_{S}(t') \otimes \hat{\rho}_{R} \) with the interaction Hamiltonian (2.87) in the generic case which has led to (2.88), it yields

\[
\hat{L}_{S}(\hat{\rho}^{S}) = \left( \hat{H}_{0}^{S}, \hat{\rho}^{S} \right) - \frac{1}{\hbar^2} \sum_{i} \int_{t_{0}}^{t} dt' \left\{ \left[ \hat{Q}_{i}, \hat{K}_{i}^{(+)}(t - t') \right] \right.
\]

\[
\left. - \left[ \hat{Q}_{i}^{\dagger}, \hat{\rho}^{S}(t - t') \hat{K}_{i}^{(-)}(t - t') \right] \right) (n_{R}^{i} + g_{i}) +
\]

\[
+ \left( \left[ \hat{Q}_{i}^{\dagger}, \hat{K}_{i}^{(-)}(t - t') \hat{\rho}^{S}(t - t') \right] - \left[ \hat{Q}_{i}, \hat{\rho}_{S}(t - t') \hat{K}_{i}^{(+)}(t - t') \right] \right) n_{i}^{R} ,
\]

(2.94)
\[ \hat{K}_i^{(+)} = e^{\frac{iS_R}{\hbar}} \hat{Q}_i e^{-\frac{iS_R}{\hbar}} e^{+\frac{iR_R}{\hbar}} \]
\[ \hat{K}_i^{(-)} = e^{\frac{iS_R}{\hbar}} \hat{Q}_i e^{-\frac{iS_R}{\hbar}} e^{-\frac{iR_R}{\hbar}} \]  \hspace{1cm} (2.96)

and \( \hat{\rho}^{iS}(t - t') \) is the system operator still in interaction picture, i.e. \( \hat{\rho}^{iS}(t - t') = \hat{U}_0^\dagger(t - t') \hat{\rho}^{S}(t') \hat{U}_0(t - t') \). The time derivative of the system operator \( \hat{O} \) becomes

\[
\frac{d}{dt} \langle \hat{O} \rangle(t) = \frac{d}{dt} tr \left\{ \hat{O} \hat{\rho}^{S}(t) \right\} = \langle \left[ \hat{O}, \hat{H}_0 \right] \rangle_S - \frac{1}{\hbar^2} \sum_i \int_{t_0}^t dt' \left\{ \langle \hat{O} [ \hat{Q}_i, \hat{K}_i^{(+)}(t - t') \hat{\rho}^{iS}(t - t')] \rangle_S - \langle \hat{O} [ \hat{Q}_i, \hat{\rho}^{iS}(t - t') \hat{K}_i^{(-)}(t - t')] \rangle_S \right\} + n_i^R + g_i \right\}.
\]

Using the unitary character of \( U_0 \) and the cyclic property of the trace we can rearrange

\[
\frac{d}{dt} \langle \hat{O} \rangle(t) = \langle \left[ \hat{O}, \hat{H}_0 \right] \rangle_S - \frac{1}{\hbar^2} \sum_i \int_{t_0}^t dt' \left\{ \langle \hat{O} [ \hat{Q}_i, \hat{K}_i^{(+)}(t - t') \hat{\rho}^{iS}(t - t')] \rangle_S - \langle \hat{O} [ \hat{Q}_i, \hat{\rho}^{iS}(t - t') \hat{K}_i^{(+)}(t - t')] \rangle_S \right\} n_i^R \right\}.
\]

If the reservoir correlation times are short, the important \( t' \) are only those close to \( t \), within this time interval the system may change rapidly — and it does — due to its unperturbed motion, but dissipative and decoherence effects may be assumed as small. This is a new form of the adiabatic limit \( \hat{\rho}^S(t') \simeq \hat{\rho}^S(t) \).

Finally, in the completed collision limit we shall have the generic form
\[
\frac{d}{dt} \langle \hat{O} \rangle(t) = \langle \left( \hat{O}, \hat{H}_0^S \right) \rangle_S - \frac{1}{\hbar^2} \sum_i \int_0^\infty du \left\{ \right.
\times \left[ (n_i^R + g_i) \left( e^{+\frac{R(t-t')}{\hbar}} \langle [\hat{O}, \hat{Q}_i^\dagger] \hat{Q}_i^\dagger(-u) \rangle_S - \right.ight.
\left. \left. e^{-\frac{R(t-t')}{\hbar}} \langle \hat{Q}_i(-u) \left[ \hat{O}, \hat{Q}_i^\dagger \right] \rangle_S \right) +
\left. + n_i^R \left( e^{-\frac{R(t-t')}{\hbar}} \langle [\hat{O}, \hat{Q}_i] \hat{Q}_i^\dagger(-u) \rangle_S - \right. \right.
\left. \left. e^{+\frac{R(t-t')}{\hbar}} \langle \hat{Q}_i(-u) \left[ \hat{O}, \hat{Q}_i^\dagger \right] \rangle_S \right) \right\}.
\]

(2.99)

If the system Hamiltonian is time independent the form is even more simple

\[
\frac{d}{dt} \langle \hat{O} \rangle(t) = \langle \left( \hat{O}, \hat{H}_0^S \right) \rangle_S - \frac{1}{\hbar^2} \sum_i \int_0^\infty du \left\{ \right.
\times \left[ (n_i^R + g_i) \left( e^{+\frac{R(t-t')}{\hbar}} \langle [\hat{O}, \hat{Q}_i^\dagger] \hat{Q}_i^\dagger(-u) \rangle_S - \right.ight.
\left. \left. e^{-\frac{R(t-t')}{\hbar}} \langle \hat{Q}_i(-u) \left[ \hat{O}, \hat{Q}_i^\dagger \right] \rangle_S \right) +
\left. + n_i^R \left( e^{-\frac{R(t-t')}{\hbar}} \langle [\hat{O}, \hat{Q}_i] \hat{Q}_i^\dagger(-u) \rangle_S - \right. \right.
\left. \left. e^{+\frac{R(t-t')}{\hbar}} \langle \hat{Q}_i(-u) \left[ \hat{O}, \hat{Q}_i^\dagger \right] \rangle_S \right) \right\}.
\]

(2.100)

We shall use the form above, but, for the sake of completeness, we shall consider the (not so common) case \( \hat{Q}_{i-1} = \hat{Q}_i \). Reconsidering the steps performed so far, in the completed collision limit, for a system Hamiltonian time independent, we have

\[
\frac{d}{dt} \langle \hat{O} \rangle(t) = \langle \left( \hat{O}, \hat{H}_0^S \right) \rangle_S - \frac{1}{\hbar^2} \sum_{i,\pm} \int_0^\infty du \left( n_i^R + \theta(\pm) \right) \times \left( e^{+\frac{R(u)}{\hbar}} \langle [\hat{O}, \hat{Q}_i] \hat{Q}_i^\dagger(-u) \rangle_S - e^{-\frac{R(u)}{\hbar}} \langle \hat{Q}_i(-u) \left[ \hat{O}, \hat{Q}_i^\dagger \right] \rangle_S \right) \]

(2.101)
2.4 Heisenberg-Langevin Approach

2.4.1 Standard quantum Langevin theory

Whenever one is dealing with a system in interaction with a huge amount of degree of freedom and, for any reason, one does not know or does not want to observe the dynamics of the exterior, a reduction problem is at hand. In the density matrix formalism of the previous chapter the system-environment relationship has been worked out in effective terms providing damping and dephasing. Another approach, complementary to the former, exists which is based on the idea from statistical mechanics that whenever an unobserved reservoir system acts on the system of interest, together with the damping also fluctuations occur. This is the aim of this approach where the same problem of eliminating the reservoir variables is worked out in a quantum operator approach. The resulting equations for the system operators include in addition to damping terms, noise operators which produce fluctuations. These equations have the same form of classical Langevin equation describing, for example, the Brownian motion of a particle suspended in a liquid. The clue idea will be to consider the unknown bath operators initial values as noise sources of a stochastic nature, stochasticity gathers by the ignorance one has on the microscopic dynamics of the whole external system. Usually the model considered is of the form of harmonic oscillators linearly coupled to a bosonic environment. The present treatment is based on [63].

Let us consider as a model the total Hamiltonian:

\[ \hat{H} = \hbar \omega_0 \left( \hat{a}^\dagger \hat{a} + \frac{1}{2} \right) + \sum_\omega \hbar \omega \left[ \hat{A}^\dagger(\omega) \hat{A}(\omega) + \frac{1}{2} \right] + \sum_\omega \hbar \left[ g(\omega) \hat{a}^\dagger \hat{A}(\omega) + g^*(\omega) \hat{A}^\dagger(\omega) \hat{a} \right], \]

(2.102)

with

\[ [\hat{a}(t), \hat{a}^\dagger(t)] = 1 = [\hat{A}(\omega,t), \hat{A}^\dagger(\omega,t)]. \]

(2.103)
The idea will be to substitute backward the bath variable into the equation of motion of the system variable and perform a Markovian approximation.

In the Heisenberg picture the equation of motion reads:

\[
\dot{\hat{a}}(t) = \frac{1}{i\hbar} [\hat{a}(t), \hat{H}] = -i\omega_0 \hat{a}(t) - i \sum_\omega g(\omega) \hat{A}(\omega, t) \tag{2.104}
\]

\[
\dot{\hat{A}}(\omega, t) = \frac{1}{i\hbar} [\hat{A}(\omega, t), \hat{H}] = -i\omega \hat{a}(\omega, t) - ig^*(\omega) \hat{\alpha}(t) \Rightarrow \\
\Rightarrow \hat{A}(\omega, t) = \hat{A}(\omega, 0) e^{-i\omega t} - ig^*(\omega) e^{-i\omega t} \int_0^t \hat{a}(t') e^{i\omega t'} dt',
\]

substituting the bath time variation of the bath operators backwards into eq. (2.104) we obtain

\[
\dot{\hat{a}}(t) = -i\omega_0 \hat{a}(t) - i \sum_\omega g(\omega) \hat{A}(\omega, 0) e^{-i\omega t} - \sum_\omega |g(\omega)|^2 \int_0^t \hat{a}(t') e^{-i\omega(t-t')} dt'. \tag{2.105}
\]

Supposing the bath spectrum \(\omega\) to have a quasi-continuum of densely distributed oscillators, we can transform from a discrete representation to a continuum representation simply introducing a density of states \(\eta(\omega)\):

\[
\sum_\omega \rightarrow \int_0^\infty \eta(\omega) d\omega.
\]

The last term in (2.105) become

\[
\sum_\omega |g(\omega)|^2 \int_0^t \hat{a}(t') e^{-i\omega(t-t')} dt' \rightarrow \int_0^t dt' \int_0^\infty d\omega \eta(\omega) |g(\omega)|^2 e^{-i\omega(t-t')} \hat{a}(t') = \\
= \int_0^t d\tau \int_{-\omega_0}^\infty d\omega' \eta(\omega' + \omega_0) |g(\omega' + \omega_0)|^2 e^{-i\omega' \tau} \hat{a}(t - \tau) e^{-i\omega_0 \tau}. \tag{2.106}
\]

Considering \(\hat{a}(t)\) to oscillate as \(e^{-i\omega_0 t}\) the last term above is a slowly varying function of \(\tau\) and one can take it out from the time integral. The other part of the integrand
in a closer look can be seen as a spectral density of the loss oscillator which, through the Wiener-Khintchine theorem, represents a memory function of the reservoir:

$$2\pi \Gamma(\tau) = \int_{-\omega_0}^{\infty} d\omega' \eta(\omega' + \omega_0)|g(\omega' + \omega_0)|^2 e^{-i\omega'\tau}. \quad (2.107)$$

The kernel $\eta(\omega' + \omega_0)|g(\omega' + \omega_0)|^2$ is a sort of spectral width gathering the memory of the interaction with the bath, i.e. the correlation time.

It is customary at this time to perform a Markov approximation:

- If $\eta(\omega' + \omega_0)|g(\omega' + \omega_0)|^2$ varies slowly on $\omega'$
- If $\omega'$ is set quite in the middle of the frequency range in order to consider $\omega_0$ as the middle point in the mean integral theorem

Then we can approximate in (2.107) the kernel as constant and equal to its value in $\omega_0$:

$$\Gamma(\tau) \simeq \eta(\omega_0)|g(\omega_0)|^2 \int_{-\omega_0}^{\infty} d\omega' e^{-i\omega'\tau} \Rightarrow$$

$$\Gamma(\tau) \simeq \eta(\omega_0)|g(\omega_0)|^2 \frac{1}{2\pi} \int_{-\omega_0}^{\infty} d\omega' e^{-i\omega'\tau} \Rightarrow$$

$$\Gamma(\tau) \simeq \eta(\omega_0)|g(\omega_0)|^2 \delta(\tau). \quad (2.108)$$

It represents the so-called Markov or memoryless approximation because in the time integration (2.105) gives:

$$\int_0^\tau \sum_\omega \left| g(\omega) \right|^2 e^{-i\omega(t-t')} \tilde{a}(t') dt' \simeq \int_0^\tau d\tau 2\pi \Gamma(\tau) e^{-i\omega_0\tau} \tilde{a}(t-\tau) \simeq 2\pi \tilde{a}(t) \int_0^\tau d\tau \Gamma(\tau) \simeq$$

$$\simeq \pi \tilde{a}(t) \eta(\omega_0)|g(\omega_0)|^2 \equiv \kappa \tilde{a}(t). \quad (2.109)$$

Finally, in equation (2.104) it reads:
\[
\dot{a} = (i\omega_0 - \kappa)\hat{a}(t) - \hat{F}(t) .
\]

(2.110)

In this equation the noise term

\[
\hat{F}(t) = i \sum_\omega g(\omega)\hat{A}(\omega,0)e^{-i\omega t}
\]

(2.111)

represents the quantum noise of the system which has in this treatment bosonic quantum statistics form the bosonic character of \(\hat{A}(\omega,0)\). The presence of the spectral width of the quantum noise \(\eta(\omega)g(\omega)\) in the damping \(\kappa\) determines a sort of Fluctuation-Dissipation theorem for the system under investigation.

### 2.4.2 Lax contribution

In the standard well-known theory of Quantum Langevin noise treatment greatly exploited in quantum optics and sketched in the previous section, 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 through the bath operators initial values. The latter, due to the impossibility to known such values, is considered as a noise source of a stochastic nature. Usually the model considered is of the form of harmonic oscillators coupled linearly to a bosonic environment. The standard statistical viewpoint is easy 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 relation for the Langevin noise sources because the bath is bosonic either.

In 1966 Melvin Lax, with clear in mind the lesson of classical statistical mechanics of brownian motion, in [64] proposed for the first time that as soon as one is left with a closed set of equations for the mean motion they can be promoted to equations for the global bare operators provided to consider additive noise sources endowed by the proper statistics due to the system dynamics. He showed that in a Markovian
environment they must fulfill generalized Einstein equations which are a sort of time
dependent non-equilibrium fluctuation dissipation theorem. The Langevin noise
source operators are such that their expectation values \( \langle \hat{F}_\mu \rangle \) vanish, but their second
order moments do not [64]. Melvin Lax noted that, although the \( \hat{F} \)'s are operators,
for most problem their reservoir averages over low-order moments and commutators
are to be known. As a consequence he restated the problem as the determination of
the moments of the noise operators (not their explicit forms) in terms of experimental
dissipation coefficients within a Markovian description. The method of attack used
has been to calculate the reservoir contribution to \( \hat{A}_\mu \) to second order in the system-
reservoir interaction. Calculate the mean moment of the noise sources \( \langle \hat{F}_\mu(t)\hat{F}_\nu(u) \rangle \)
to second order in the interaction too. By comparing the coefficients in these two
calculations he arrived at a fluctuation-dissipation theorem valid for nonequilibrium
situations.

If \( \hat{a} = \{\hat{a}_1, \hat{a}_2, \cdots \} \) is a set of system operators, and

\[
\frac{d \langle \hat{a}_\mu \rangle}{dt} = \langle \hat{A}_\mu(\hat{a}) \rangle
\]

(2.112)

are the correct equations for the mean, then one can show that the equations

\[
\frac{d\hat{a}_\mu}{dt} = \hat{A}_\mu(\hat{a}) + \hat{F}_\mu(\hat{a},t)
\]

(2.113)

are a valid set of equations of motion for the operators provided the additive oper-
ators \( \hat{F} \)'s to be endowed with the correct statistical properties to be determined for
the motion itself.

The assumption that the system is Markovian can be set rigorously in the form

\[
\hat{F}_\mu(s) \text{ is independent of } \hat{a}_\nu(t) \text{ if } s > t
\]

(2.114)

and in particular they must commute. If not obeyed the statistic of the random
forces \( \hat{F} \) would depend on the past history and the system would acquire a memory,
in contrast to the Markovian or memoryless behaviour. However, for the proof, the
only thing mathematically needed is the above condition somewhat relaxed because the only thing used is the lack of correlation between the random force at a time and an arbitrary system operator \( C(\hat{a}) \) at an earlier time:

\[
\langle \hat{F}_\mu(s)C(\hat{a}(t)) \rangle = \langle C(\hat{a}(t))\hat{F}_\mu(s) \rangle = 0 \quad \text{if} \quad s > t .
\] (2.115)

In order to obtain the relations, it is sufficient to start from the algebraic identity

\[
\hat{a}_\mu(t + \Delta t)\hat{a}_\nu(t + \Delta t) - \hat{a}_\mu(t)\hat{a}_\nu(t) = \\
= \Delta \hat{a}_\mu \Delta \hat{a}_\nu + \Delta \hat{a}_\mu \hat{a}_\nu + \hat{a}_\mu \Delta \hat{a}_\nu ,
\] (2.116)

with

\[
\Delta \hat{a}_\nu = \hat{a}_\nu(t + \Delta t) - \hat{a}_\nu(t) \\
\Delta \hat{a}_\nu \simeq \hat{A}_\nu \Delta t + \int_t^{t+\Delta t} \hat{F}_\nu(s)ds .
\] (2.117)

Dividing by \( \Delta t \), in the limit \( \Delta t \to 0 \) one can obtain the equation of motion for the mean of two operators:

\[
\frac{d \langle \hat{a}_\mu(t)\hat{a}_\nu(t) \rangle}{dt} = 2 \langle D_{\mu\nu} \rangle + \langle \hat{A}_\mu \hat{a}_\nu \rangle + \langle \hat{a}_\mu \hat{A}_\nu \rangle .
\] (2.118)

In the following mnemonic form it acquires a highly evocative meaning:

\[
2 \langle D_{\mu\nu} \rangle = -\{d(\hat{a}_\mu)/dt\}\hat{A}_\nu - \{\hat{A}_\mu\{d(\hat{a}_\nu)/dt\}\} + \{d(\hat{a}_\mu(t)\hat{a}_\nu(t))/dt\} ,
\] (2.119)

where \( \{d(\hat{a}_\mu)/dt\} \equiv d(\hat{a}_\mu)/dt - \hat{F}_\mu \equiv \hat{A}_\mu .\)

They are intimately linked up with the global dissipation and in a Markovian environment they take the form:

\[
\langle \hat{F}_\mu(t)\hat{F}_\nu(u) \rangle = 2 \langle D_{\mu\nu} \rangle \delta(t-u) .
\] (2.120)
Equation (2.120) is an (exact) time dependent Einstein equation representing a fluctuation-dissipation relation valid for nonequilibrium situations, it witnesses the fundamental correspondence between dissipation and noise in an open system. Here we may divide the average process into two subsequent steps: a single bracket \( \langle \rangle \) denoting reservoir average of a global operator, then \( \langle \hat{a} \rangle \) is still a system operator averaged over the bath degrees of freedom, and an additional bracket for the averaging over the system itself, thus \( \langle \langle \hat{a} \rangle \rangle \) is a \( C \)-number, i.e. an physical quantity. In this guise the single \( \langle \rangle \) can be considered even reservoir averages, thus, in general, \( \langle D_{\mu\nu} \rangle \) is 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. Operating in this way the resulting "fluctuation-dissipation" relations between \( D_{\mu\nu} \) and the reservoir contributions are 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. Higher order diffusion coefficients may be determined by a straightforward generalization of (2.119). However higher order moments of the Langevin forces would be related in a more complicated way to system diffusion constant [64].
Part II

Non-classical properties of light in the strong-coupling regime
Chapter 3

Atom optics and nonlinear coherent optical response in semiconductor microcavities

3.1 Incoherent dynamics and parametric processes

Entanglement is one of the key features of quantum information and communication technology [10]. Parametric down-conversion is the most frequently used method to generate highly entangled pairs of photons for quantum-optics applications, such as quantum cryptography and quantum teleportation. Rapid development in the field of quantum information requires monolithic, compact sources of nonclassical photon states enabling efficient coupling into optical fibres and possibly electrical injection. Semiconductor-based sources of entangled photons would therefore be advantageous for practical quantum technologies. Moreover semiconductors can be structured on a nanometer scale, and thus one may produce materials with tailored properties realizing a wide variety of physically distinct situations. However semiconductor heterostructures constitute a complex interacting environment involving charge, spin,
3 – Atom optics and nonlinear coherent optical response in semiconductor microcavities

and lattice degrees of freedom, hence suited to serve as prototype systems where quantum-mechanical properties of many interacting particles far away from equilibrium can be studied in a controlled fashion [41]. It has been demonstrated that very large $\chi^{(3)}$ resonant polaritonic nonlinearities in wide-gap semiconductors and in semiconductor microcavities can be used to achieve parametric emission [65, 12].

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 [65], the possibility of generating entangled photons by these processes was theoretically pointed out only lately [20]. This result was based on a microscopic quantum theory of the nonlinear optical response of interacting electron systems relying on the dynamics controlled truncation scheme [59] extended to include light quantization [66, 47, 19]. The above theoretical framework was also applied to the analysis of polariton parametric emission in semiconductor microcavities (SMCs) [66, 47]. A SMC is a photonic structure designed to enhance light-matter interactions. The strong light-matter interaction in these systems gives rise to cavity polaritons which are hybrid quasiparticles consisting of a superposition of cavity photons and quantum well excitons [11]. Demonstrations of parametric amplification and parametric emission in SMCs [12, 13, 14], together with the possibility of ultrafast optical manipulation and ease of integration of these microdevices, have increased the interest on the possible realization of nonclassical cavity-polariton states [15, 16, 17, 18, 19]. In 2004, 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 [23]. Short-wavelength entangled photons are desirable for a number of applications as generation of further entanglement between three or four photons. In 2005 an experiment probing quantum correlations of (parametrically emitted) cavity
polaritons by exploiting quantum complementarity has been proposed and realized [17]. Specifically, it has been shown that polaritons in two distinct idler modes interfere if and only if they share the same signal mode so that which-way information cannot be gathered, according to Bohr’s quantum complementarity principle. In 2006 a promising low-threshold parametric oscillation in vertical triple SMCs with signal, pump and idler waves propagating along the vertical direction of the nanostructure has been demonstrated [68].

The crucial role of many-particle Coulomb correlations in semiconductors marks a profound difference from dilute atomic systems, where the optical response is well described by independent transitions between atomic levels, and the non-linear dynamics is governed only by saturation effects. In planar SMCs, thanks to their mutual Coulomb interaction, pump polaritons generated by resonant optical pumping may scatter into pairs of polaritons (signal and idler) [20, 12, 21], they are determined by the two customary energy and wave vector conservation conditions $2k_p = k_s + k_i$ and $2E_{k_p} = E_{k_s} + E_{k_i}$ depicting an eight-shaped curve in momentum space. At low pump intensities they are expected to undergo a spontaneous parametric process driven by vacuum-fluctuation, whereas at moderate intensities they display self-stimulation and oscillation [12]. However they are real electronic excitation 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 noise and dephasing induced by electron-phonon interaction [42].

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 \[22, 21, 18, 16, 14\]. According to these theoretical calculations, parametric processes are already visible in principle as well as the pump is switched on even at very low excitation intensities. On the contrary, in experiments, what really dominates the light emission at low pump intensities is the photoluminescence (PL) due to the incoherent dynamics of the single scattering events driven by the pump itself. Only once the pumping become sufficient the parametric processes start to reveal themselves and to take over pump-induced PL as well. Indeed, usually, parametric emission and standard pump-induced PL cohabit as shown by experiments at low and intermediate excitation density \[14\]. Moreover, in order to address quantum coherence properties and entanglement \[23\] the preferred experimental situations are those of few-particles 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. Thus, a microscopic analysis able to account for parametric emission and pump-induced PL on an equal footing is highly desired in order to make quantitative comparison 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 the unwanted detrimental contributions.

The dynamics controlled truncation scheme (DCTS) provides a (widely adopted) starting point for the microscopic theory of light-matter interaction effects beyond mean-field \[41, 59\], supplying a consistent and precise way to stop the infinite hierarchy of higher-order correlations which always appears in the microscopic approaches of manybody interacting systems. In 1996 the DCTS was extended in order to include in the description the quantization of the electromagnetic field \[20\]. This
extension has been applied to the study of quantum optical phenomena in semiconductors as polariton entanglement [19]. However in these works damping has been considered only at a phenomenological level.

In this chapter we shall present a novel approach based on a DCTS-nonequilibrium quantum Langevin description of the open system in interaction with its surroundings we published in Ref. [25]. In order to make our presentation clearer in Chapter 4.1 we shall present in great details the approach to polaritons in QW microcavities presented in Ref. [20], afterwards from Chapter 4.2 our original approach will be addressed. By means of a sort of microscopic quantum fluctuation-dissipation theorem it enables us to include on an equal footing the microscopic description of the scattering channels competing with the coherent parametric phenomena the optical pump induces. We shall apply our method in order to perform a realistic description of light emission taking into account nonlinear parametric interactions, light quantization, cavity losses and polariton-phonon interaction. We shall present a DCTS theory for interacting polaritons, the focus will be on the nonlinear part in order to model coherent optical parametric ($\chi^{(3)}$) processes and the damping is included only phenomenologically. The subsequent section will be devoted to the microscopic calculation of phonon-induced scattering and polariton PL within a second order Born-Markov approach. Then we shall present a quantum Langevin description of parametric emission including incoherent effects and particular attention will be devoted to the case of single pump feed.
Chapter 4

Coherent optical response beyond mean-field in the 1S exciton sector

Semiconductor planar microcavities polaritons are hybrid quasi-particles originating from the strong-coupling between cavity photons and two-dimensional excitons in an embedded quantum well. Thanks to their mutual Coulomb interaction, pump polaritons generated by resonant optical pumping can scatter into pairs of polaritons (signal and idler), in the low excitation limit they are expected to undergo a spontaneous parametric process driven by vacuum-fluctuation, whereas at moderate excitation intensities it displays self-stimulation. 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.

Our aim is to build up a clear and versatile theoretical tool able to focus on non-linear coherent optical response and incoherent contributions on an equal footing. With clear in mind the lesson gather from quantum optics we consider a Quantum Langevin approach to the problem; in the subsequent sections all the steps pursued
in order to obtain our goal will be touched in detail. This enables the microscopic analysis of the influence of decoherence and noise on the polariton quantum correlations originating from nonlinear optical processes.

4.1 The coupled system

The system we have in mind is a quantum well grown inside a semiconductor planar Fabry-Perot resonator. Using a perturbation series expansion in the exciting field a systematic theory of Coulomb interaction effects in the nonlinear optical response in semiconductors is presented. Pushing forward the calculation up to the third order in the electric field all the relevant coherent dynamics may be set within the 1S exciton sector, we end up with a phase-space filling correction, a mean-field exciton-exciton interaction and a 4-particle correlation terms.

In order to set the notation we are about to use throughout the calculation we begin with saying that \( |0\rangle \) denotes the trivial ground state with no eh-pairs present. The one-exciton subspace is the closure of the linear span of \( |E_{n,k}^{(1)}\rangle \equiv |1nk\rangle \) with energy \( \hbar \omega_{1,n,k} \), it is worth noticing that this way both bound and scattering states are included in \( n \), meanwhile \( k = (\sigma, k) \), the polarization index \( \sigma \) stands to remember which specific optical transition is required in order to excite the eh-pair and hence to create the exciton, if optically active this correspond to the helicity of the light involved, \( k \) is the center of mass wavevector. The next relevant subspace is the biexciton one corresponding to all the 4-particles aggregates. It is built up from the complete basis \( |E_{m}^{(2)}\rangle \) with \( w_{2,m} \) where \( m \) represents the suitable collection of quantum numbers needed to specify the bound as well the unbound states. A relevant point is linked up with this latter subspace. Even though not all the biexciton states are computed due to the many-body nature of the problem, we will be able to keep as long as possible all the biexcitons inside the calculation because there
will be situations where they arise as intermediate states and can be resummed up by virtue of the completeness theorem. It may be worth noting that it is usually a very good approximation at low temperatures to represent the ground state of an intrinsic semiconductor by a single Slater determinant corresponding to filled valence and empty conduction bands. The most important consequence of such approximation is to neglect of the eh pair fluctuation which affect the ground state and the dielectric screening of the Coulomb interaction. To be specific the ground state of the semiconductor is defined as the vacuum state with respect to the exciton annihilation

$$\hat{B}_{n\sigma} |0\rangle = 0, \hat{B}_{nk} = \sum_{k'} \Phi_{n\sigma k'}^* \hat{a}_{h,-k'+\eta_h k} \hat{a}_{c,k'+\eta_c k}$$

(4.1)

which means that no eh-pairs are presented in the semiconductor ground state. The dielectric screening is approximately accounted for by the static dielectric constant of the semiconductor. The eigenstates of the Hamiltonian $\hat{H}_{ph}$ of the cavity modes are written as $|n,\lambda\rangle$ where $n$ stands for the total number of photons in the state and $\lambda = (k_1,\sigma_1; \ldots; k_n,\sigma_n)$ specifies wave vector and polarization $\sigma$ of each photon. For the absence of any interaction mechanism among them it is clear that it is nothing but a product state. We mean that from standard theory is known that any tensor product is specifically the normed closure of the linear span of the product states including this way all the possible vector belonging to such a Hilbert space, this is the case of exciton; as for the photon geometrically speaking the procedure is the same, but the eigenstates are precisely product states, it seems a pointless discussion but the property to be a product state yields the immediate result that $(n = \sum_i n_i, m = \sum_j m_j)$

$$|n,\lambda\rangle = \bigotimes_i |n_i, (k_i,\sigma_i)\rangle = \bigotimes_i \frac{\hat{a}^\dagger_{k_i,\sigma_i}^{(n_i)}}{\sqrt{(n_i + 1)!}} |0\rangle$$

$$|n\lambda\rangle \langle m\mu| = \bigotimes_{i,j} \frac{\hat{a}_{k_i,\sigma_i}^{(n_i)}}{\sqrt{(n_i + 1)!}} \frac{\hat{a}_{k_j,\sigma_j}^{(m_j)}}{\sqrt{m_j!}} |0\rangle \langle 0|,$$

(4.2)
loosely written also as simple product.

The two Hamiltonians are

\[ \hat{H}_0 = \hat{H}_{\text{exc}} + \hat{H}_{\text{cav}} = \sum_{N\alpha} \hbar \omega_{N\alpha} \left| E_{N\alpha} \right\rangle \langle E_{N\alpha} \right| + \sum_{k} \hbar \omega_{k} \hat{a}^\dagger_{k} \hat{a}_{k}. \] (4.3)

The coupling of the electron system and the cavity modes is given in the usual rotating wave approximation

\[ \hat{H}_{\text{cav-exc}} = -\sum_{nk} V_{nk} \hat{a}^\dagger_{k} \hat{B}_{nk} + \text{H.c.} \] (4.4)

with the operator \( \hat{a}^\dagger_{k} \) which creates a photon state with \( k = (\sigma, \mathbf{k}) \) and energy \( \hbar \omega_{k} = \hbar (\omega_{\text{exc}}^{2} + v^{2}|\mathbf{k}|^{2})^{1/2} \), \( v \) being the velocity of light inside the cavity, \( \hat{B}^\dagger_{n,k} \) creates an exciton state with the same wave vector and polarization \( k \) and energy \( \hbar \omega_{1,n,k} \). \( V_{n,k} \) is the photon-exciton coupling coefficient enhanced by the presence of the cavity [54], we shall set \( V_{n,k} = \tilde{V}_{\mathbf{k}}(0) \) in order to be a little more free when explicitly calculating the contributions, physically speaking we want to set our attention on the polariton resonance, but we know that in order to describe properly light-matter interaction through its polarization field [48] one must include the notion of the shape of the exciton envelope wave function [47, 24].

We shall treat the cavity field in the quasi-mode approximation, that is to say we shall quantize the field as the mirror were perfect and subsequently we shall couple the cavity with a statistical reservoir of a continuum of external modes. This way on an equal footing we shall provide the coherent driving mechanism as the ensemble average, important at a coherent (first order in the interaction) level, as well as the radiative damping channel (at work within a second order Born-Markov description).

\[ \hat{H}_{qm} = i\hbar \sum_{k\|} \int d\omega g_{k\|}(\omega) \hat{a}^\dagger_{k\|} \hat{E}^{(-)}_{k\|}(\omega,t) + \text{H.c.} \] (4.5)
In passing form the air to the sample we change from a 3D to a 2D quantization, it means that in the coupling once either \((\mathbf{k}^{\parallel}, k_z)\) or \((\mathbf{k}^{\parallel}, \omega)\) is chosen the third follow consistently. We have chosen the latter for simplicity in dealing with the Markov machinery. In the Hamiltonian \(g_{\mathbf{k}^{\parallel}}(\omega)\) is the coupling coefficient, a sort of optical matrix element, \(\hat{E}^{(-)}_{\mathbf{k}^{\parallel}}(\omega,t)\) and \(\hat{E}^{(+)}_{\mathbf{k}^{\parallel}}(\omega,t)\) are the two propagating normal modes of the external light. Modeling 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). It will be nothing but the non zero expectation value of the ensemble itself giving a non zero contribution on the 1\(^{st}\) order. It will be analyzed in detail in section 5.2, for the time being we shall model the coherent input as \([20]\). In 5.2 we shall set the conditions one must fulfill in order to model the pump term as we are about to do.

\[
\hat{H}_p = i t_c \sum_k (E_k \hat{a}_k^{\dagger} - E_k^{\star} \hat{a}_k) .
\]  

(4.6)

The idea is not to use a density matrix approach, but to construct expectation values of all the quantities at play. The dynamics is described by so-called generalized Hubbard operators (nothing but general projectors):

\[
\hat{X}_{N,\alpha;M,\beta} \hat{Y}_{n,\lambda;m,\mu}
\]

(4.7)

where

\[
\hat{X}_{N,\alpha;M,\beta} = | E_{N,\alpha} \rangle \langle E_{M,\beta} |
\]

\[
\hat{Y}_{n,\lambda;m,\mu} = | n,\lambda \rangle \langle m,\mu |
\]

The fundamental point in the whole analysis is that due to the form of the interaction Hamiltonian \(\hat{H}_I\) we can use the so-called Dynamics Controlled Truncation Scheme,
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 holds. Expressed in our notation it reads [20]:

\[
\langle \hat{X}_{N,\alpha;M,\beta}\hat{Y}_{n,\lambda;m,\mu} \rangle = \sum_{i=0}^{i_0} \langle \hat{X}_{N,\alpha;M,\beta}\hat{Y}_{n,\lambda;m,\mu} \rangle^{(N+M+n+m+2i)} + \mathcal{O}(E^{(N+M+n+m+2i_0+2)}) \tag{4.8}
\]

the expectation value of a zero to \textbf{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 general ingredient of interest in the present discussion is

\[
\hat{X}_{N,\alpha;M,\beta}\hat{Y}_{n,\lambda;m,\mu} , \tag{4.9}
\]

the excitons and photons operators can be expressed as

\[
\hat{a}_k = \hat{Y}_{0,1k} + \sum_{n \geq 1} \langle n\gamma | \hat{a}_k | (n + 1)(\gamma,k) \rangle \hat{Y}_{n\gamma;(n+1)(\gamma,k)} = \hat{Y}_{0,1k} + \sum_{n \geq 1} \sqrt{n + 1}\hat{Y}_{n\gamma;(n+1)(\gamma,k)}
\]

\[
\hat{B}_{nk} = \hat{X}_{0,1nk} + \sum_{N \geq 1,\alpha\beta} \langle N\alpha | \hat{B}_{nk} | (N + 1)\beta \rangle \hat{X}_{n\alpha;(N+1)\beta} . \tag{4.10}
\]

The equation of motion for (4.9), under the Hamiltonians \(\hat{H}_{\text{exc}}, \hat{H}_{\text{cav}}, \hat{H}_{\text{cav-exc}}, \hat{H}_{p}\) reads:
\[
\frac{d}{dt} (\hat{X}_{N\alpha;M\beta} \hat{Y}_{n\lambda;m\mu}) = -i(\omega_{M\beta} - \omega_{N\alpha} + \sum_{i=1}^{m} \omega_{k_i} - \sum_{j=1}^{n} \omega_{\ell_j}) (\hat{X}_{N\alpha;M\beta} \hat{Y}_{n\lambda;m\mu}) +
\]
\[
+ \hat{X}_{N\alpha;M\beta} (\delta_{m,1} t_0 \frac{E_m}{\hbar} \hat{Y}_{n\lambda;0} + \delta_{n,1} t_0 \frac{E_n}{\hbar} \hat{Y}_{0;m\mu}) - \hat{X}_{N\alpha;M\beta} \sum_{k} t_c (\delta_{m,0} \frac{E_k}{\hbar} \hat{Y}_{n\lambda;1k} + \delta_{n,0} \frac{E_k}{\hbar} \hat{Y}_{1k;m\mu}) +
\]
\[
+ \sum_{k\nu} t_c \hat{X}_{N\alpha;M\beta} \left[ \Theta(m - 2) \langle m\mu | \hat{a}_k^\dagger | (m - 1)\nu \rangle \frac{E_k}{\hbar} \hat{Y}_{n\lambda;m(1 - 1)\nu} + \right.
\]
\[
\left. \Theta(n - 2) \langle (n - 1)\nu | \hat{a}_k | n\lambda \rangle \frac{E_k}{\hbar} \hat{Y}_{(n-1)\nu;m\mu} - \right.
\]
\[
\left. \Theta(m - 1) \langle m\mu | \hat{a}_k^\dagger | (m + 1)\nu \rangle \frac{E_k}{\hbar} \hat{Y}_{n\lambda;m(1 + 1)\nu} - \right.
\]
\[
\left. \Theta(n - 1) \langle (n + 1)\nu | \hat{a}_k | n\lambda \rangle \frac{E_k}{\hbar} \hat{Y}_{(n+1)\nu;m\mu} \right] +
\]
\[
\frac{i}{\hbar} \delta_{M,0} \delta_{\beta,0} \delta_{m,1} \sum_{\tilde{n}} V_{\tilde{n}\mu} \hat{X}_{N\alpha;\tilde{n}\mu} \hat{Y}_{n\lambda;0} - \frac{i}{\hbar} \delta_{N,0} \delta_{\alpha,0} \delta_{n,1} \sum_{\hat{n}} V_{\hat{n}\lambda} \hat{X}_{\tilde{1n}\lambda} \hat{Y}_{0;m\mu} -
\]
\[
- \frac{i}{\hbar} \delta_{N,1} \delta_{\alpha,0} \delta_{n,0} V\hat{n}\mu \hat{X}_{\tilde{0};M\beta} \hat{Y}_{1k;\mu m} + \frac{i}{\hbar} \delta_{M,1} \delta_{m,0} \delta_{\beta,0} V\hat{n}\lambda \hat{X}_{N\alpha;0} \hat{Y}_{n\lambda;1k} +
\]
\[
+ \frac{i}{\hbar} \delta_{m,1} \Theta(M - 1) \sum_{\tilde{n}\delta} V\tilde{n}\mu | M\beta \rangle \hat{B}_{\tilde{n}\mu} | (M + 1)\delta \rangle \hat{X}_{N\alpha;\tilde{n}\delta} \hat{Y}_{n\lambda;0} -
\]
\[
- \frac{i}{\hbar} \delta_{n,1} \Theta(N - 1) \sum_{\tilde{n}\eta} V_{\tilde{n}\lambda} \langle (N + 1)\eta | \hat{B}_{\tilde{n}\lambda} | N\alpha \rangle \hat{X}_{\tilde{n}(N+1)\eta;M\beta} \hat{Y}_{0;m\mu} -
\]
\[
- \frac{i}{\hbar} \delta_{n,0} \delta_{\alpha,0} \Theta(N - 2) \sum_{\tilde{n}\kappa\eta} V_A_{\tilde{n}\kappa} \langle (N - 1)\eta | \hat{B}_{\tilde{n}\kappa} | N\alpha \rangle \hat{X}_{\tilde{n}(N-1)\eta;M\beta} \hat{Y}_{1k;m\mu} +
\]
\[
+ \frac{i}{\hbar} \delta_{m,0} \delta_{\mu,0} \Theta(M - 2) \sum_{\tilde{n}\delta\kappa} V_A_{\tilde{n}\kappa} \langle m\mu | \hat{B}_{\tilde{n}\kappa} | (M - 1)\delta \rangle \hat{X}_{N\alpha;\tilde{n}\delta\kappa} \hat{Y}_{n\lambda;1k} +
\]
\[
+ \frac{i}{\hbar} \delta_{M,0} \delta_{\beta,0} \Theta(m - 2) \sum_{\tilde{n}\kappa\nu} V_{\tilde{n}\nu} \langle m\mu | \hat{a}_k^\dagger | (m - 1)\nu \rangle \hat{X}_{\tilde{n}1k\mu;M\beta} \hat{Y}_{n\lambda;1k\nu} -
\]
\[
- \frac{i}{\hbar} \delta_{N,0} \delta_{\alpha,0} \Theta(n - 2) \sum_{\tilde{n}\gamma\kappa} V_{\tilde{n}\kappa} \langle (n - 1)\gamma | \hat{a}_k | n\lambda \rangle \hat{X}_{\tilde{n}1k\mu;M\beta} \hat{Y}_{n\lambda;1k\nu} -
\]
\[
- \frac{i}{\hbar} \delta_{N,1} \Theta(n - 1) V\hat{n}\alpha \sum_{\gamma} \langle (n + 1)\gamma | \hat{a}_k^\dagger | n\lambda \rangle \hat{X}_{\tilde{0};M\beta} \hat{Y}_{(n+1)\gamma;m\mu} +
\]
\[
+ \frac{i}{\hbar} \delta_{M,1} \Theta(m - 1) V\beta \sum_{\nu} \langle m\mu | \hat{a}_k^\dagger | (m + 1)\nu \rangle \hat{X}_{N\alpha;0} \hat{Y}_{n\lambda;1k\nu} +
\]
\[
+ \frac{i}{\hbar} \sum_{\tilde{n}k} \sum_{\nu \delta} \left[ V_{\tilde{n}k}^* \left( \Theta(M - 1) \Theta(m - 2) \langle M \beta \mid \hat{B}_{\tilde{n}k} \mid (M + 1) \delta \rangle \right. \right.
\]

\[
\left. \langle m \mu \mid \hat{a}_k^\dagger \mid (m - 1) \nu \rangle \hat{X}_{N \alpha ; (M + 1) \delta} \hat{Y}_{n \lambda ; (m - 1) \nu} - \right.
\]

\[
- \Theta(N - 2) \Theta(n - 1) \langle (N - 1) \delta \mid \hat{B}_{\tilde{n}k} \mid N \alpha \rangle \right.
\]

\[
\left. \langle (n + 1) \nu \mid \hat{a}_k^\dagger \mid n \lambda \rangle \hat{X}_{(N - 1) \delta ; M \beta} \hat{Y}_{(n + 1) \nu ; m \mu} \right. -
\]

\[
- V_{\tilde{n}k} \left( \Theta(N - 1) \Theta(n - 2) \langle (N + 1) \delta \mid \hat{B}_{\tilde{n}k}^\dagger \mid N \alpha \rangle \right.
\]

\[
\left. \langle (n - 1) \nu \mid \hat{a}_k \mid n \lambda \rangle \hat{X}_{(N + 1) \delta ; M \beta} \hat{Y}_{(n - 1) \nu ; m \mu} - \right.
\]

\[
- \Theta(M - 2) \Theta(m - 1) \langle M \beta \mid \hat{B}_{\tilde{n}k}^\dagger \mid (M - 1) \delta \rangle \right.
\]

\[
\left. \langle m \mu \mid \hat{a}_k \mid (m + 1) \nu \rangle \hat{X}_{N \alpha ; (M - 1) \delta} \hat{Y}_{n \lambda ; (m + 1) \nu} \right\} .
\] (4.11)

Here \( \Theta(x) \) is the Heaviside function equal to 1 for positive argument and zero otherwise.

The linear dynamics is very easy \( \langle \hat{a}_k \rangle^{(1)} = \langle \hat{X}_{0,0} \hat{Y}_{1,nk} \rangle^{(1)} \) and \( \langle \hat{B}_{nk} \rangle^{(1)} = \langle \hat{X}_{0,1nk} \hat{Y}_{0,0} \rangle^{(1)} \):

\[
\frac{d}{dt} \langle \hat{a}_k \rangle^{(1)} = -i \omega_k \langle \hat{a}_k \rangle^{(1)} + i \sum_{\tilde{n}} \frac{V_{nk}}{\hbar} \langle \hat{B}_{nk} \rangle^{(1)} + t_c \frac{E_k}{\hbar}
\] (4.12)

\[
\frac{d}{dt} \langle \hat{B}_{nk} \rangle^{(1)} = -i \omega_{1nk} \langle \hat{B}_{nk} \rangle^{(1)} + i \frac{V_{nk}}{\hbar} \langle \hat{a}_k \rangle^{(1)} .
\] (4.13)

The dynamics up to the third order is a little bit more complex, we shall make extensively use of (4.11) (in the following the suffix \( +(\cdot) \) stands for “up to” that order).

\[
\langle \hat{B}_{nk} \rangle^{+(3)} = \langle \hat{X}_{0,1nk} \hat{Y}_{0,0} \rangle^{(1)} + \sum_\gamma \langle \hat{X}_{0,1nk} \hat{Y}_{1 \gamma ; 1 \gamma} \rangle^{(3)} + \sum_{\alpha \beta} \langle 1 \alpha \mid \hat{B}_{nk} \mid 2 \beta \rangle \langle \hat{X}_{1 \alpha ; 2 \beta} \hat{Y}_{0,0} \rangle^{(3)} ,
\] (4.14)
\begin{equation}
\langle \hat{a}_k \rangle^{(3)} = \langle \hat{X}_{0;0}\hat{Y}_{0;1k} \rangle^{(1)} + \sum_\beta \langle \hat{X}_{1;1\beta}\hat{Y}_{0;1nk} \rangle^{(3)} + \sqrt{2} \sum_\gamma \langle \hat{X}_{0;0}\hat{Y}_{1;2(\gamma,k)} \rangle^{(3)} .
\end{equation}

With a bit of algebra we obtain

\begin{equation}
\frac{d}{dt} \langle \hat{a}_k \rangle^{(3)} = -i\omega_k \langle \hat{a}_k \rangle^{(3)} + i \sum_n \frac{V_{nk}}{\hbar} \langle \hat{B}_{nk} \rangle^{(3)} + i\epsilon_t \frac{E_k}{\hbar} ,
\end{equation}

\begin{equation}
\frac{d}{dt} \langle \hat{B}_{nk} \rangle^{(3)} = -\tilde{\omega}_{1nk} \langle \hat{B}_{nk} \rangle^{(3)} + i \frac{V_{nk}}{\hbar} \langle \hat{a}_k \rangle^{(3)} +
\sum_{\tilde{n}k} \left[ i \sum_{n'k',\beta} V_{n'k'} \langle 1\tilde{n}\tilde{k} | [\hat{B}_{nk},\hat{B}_{n'k'}^{\dagger}] - \delta_{n'(n'k')/(nk)} | 1\beta \rangle \langle \hat{X}_{1\tilde{n}\tilde{k};1\beta}\hat{Y}_{0;1k'} \rangle^{(3)} -
- i \sum_\beta \langle \tilde{\omega}_{2\beta} - \tilde{\omega}_{1\tilde{n}k} - \tilde{\omega}_{1nk} \rangle^{(1)} \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{equation}

where for brevity we have use in the nonlinear part the $\beta$ index once as an exciton label, whereas in the last line it is a biexciton index. For later convenience we have introduce phenomenological damping terms, e.g. $\tilde{\omega}_{1nk} = \Gamma_x + i\omega_{1nk}$ and $\tilde{\omega}_{2\beta} = \Gamma_x + i\omega_{2\beta}$. The contribution to the equation of motion for the biexciton subspace is

\begin{equation}
\frac{d}{dt} \langle \hat{X}_{0;2\beta} \rangle^{(2)} = -(\Gamma_{2\beta} + i\omega_{2\beta}^{x}) \langle \hat{X}_{0;2\beta} \rangle^{(2)} +
+i \sum_{n'k',n''k''} V_{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{equation}

Thanks to the fact we want to treat coherent optical processes it is possible to manipulate further the parametric contributions under two assumptions. We are addressing a coherent optical response, thus we may consider that a coherent pumping mainly generates \textit{coherent} parametric processes, as a consequence the dominant contribution of the biexcitonic sector on the third-order nonlinear response would
be of a coherent type; we mean that we shall safely consider \( \langle \hat{X}_{1\tilde{n}_k;0} \rangle^{(1)} \langle \hat{X}_{0;2\beta} \rangle^{(2)} I_{\text{cav}} \). In addition we make the stronger approximation stating that also for the other contribution in Eq. (4.17) we may write

\[
\langle \hat{X}_{1\tilde{n}_k;0} \rangle^{(1)} \langle \hat{X}_{0;2\beta} \rangle^{(2)} I_{\text{cav}}.
\]

In addition we make the stronger approximation stating that also for the other contribution in Eq. (4.17) we may write

\[
\langle \hat{X}_{1\tilde{n}_k;0} \rangle^{(1)} \langle \hat{X}_{0;2\beta} \rangle^{(2)} I_{\text{cav}}.
\]

This is a very strong assumption for it neglects higher order quantum optical correlation effects between the electron system and the cavity modes leading to a renormalization of the biexcitonic dynamics with intriguing physical perspectives. However for extended systems, like QWs in planar microcavities, these are effects in most cases of negligible impact, on the contrary in confined geometries such as cavity embedded quantum dots they could given significant contributions. In the end, within such a coherent limit we are able to describe the biexcitonic contribution effectively as an exciton correlation.

The first consequence is immediately on eq. (4.18):

\[
\frac{d}{dt} \langle \hat{X}_{0;2\beta} \rangle^{(2)} = -(\Gamma_{2\beta} + i\omega_{2\beta}) \langle \hat{X}_{0;2\beta} \rangle^{(2)} + i \sum_{n'k',n''k''} V_{n'k'} \langle 2\beta | \hat{B}_{n'k'}^{\dagger} | 1n''k'' \rangle \langle \hat{a}_{k'}^{(1)} \rangle \langle \hat{B}_{n''k''} \rangle^{(1)} + \frac{i}{\hbar} \sum_{nk} \langle \hat{B}_{nk} \rangle^{(1)} R_{nk;\tilde{n}_k}^{(2)}, \tag{4.19}
\]

The nonlinear \((\chi^{(3)})\) dynamics becomes:

\[
\frac{d}{dt} \langle \hat{B}_{nk} \rangle^{(3)} = -\omega_{nk} \langle \hat{B}_{nk} \rangle^{(3)} + \frac{V_{nk}}{\hbar} \langle \hat{a}_{k} \rangle^{(3)} - i \sum_{\tilde{n}_k} \langle \hat{B}_{\tilde{n}_k} \rangle^{(1)} c_{nk;\tilde{n}_k;\beta}^{(1)} \langle \hat{X}_{0;2\beta} \rangle^{(2)}, \tag{4.20}
\]

where

\[
R_{nk;\tilde{n}_k}^{(2)} = \sum_{n'k',n''k''} C_{nk;\tilde{n}_k}^{n'k',n''k''} \langle \hat{B}_{n''k''} \rangle^{(1)} \langle \hat{a}_{k'}^{(1)} \rangle + \sum_{\beta} c_{nk;\tilde{n}_k;\beta}^{(1)} \langle \hat{X}_{0;2\beta} \rangle^{(2)}, \tag{4.21}
\]

with

\[
C_{nk;\tilde{n}_k}^{n'k',n''k''} = V_{n'k'} \langle 1\tilde{n}_k | \delta_{(n'k'):(nk)} - [\hat{B}_{nk}, \hat{B}_{n'k'}^{\dagger}] | 1n''k'' \rangle \tag{4.22}
\]

\[
c_{nk;\tilde{n}_k;\beta}^{(1)} = \hbar (\omega_{2\beta} - \omega_{1\tilde{n}_k} - \omega_{1nk}) \langle 1\tilde{n}_k | \hat{B}_{nk} | 2\beta \rangle. \tag{4.23}
\]
This equation is analogous to the corresponding equation describing the semiclassical (quantized electron system, classical light field) coherent $\chi^{(3)}$ response in a QW [24], the main difference being that here the (intracavity) light field is regarded not as a driving external source but as a dynamical field [66]. This close correspondence for the dynamics of expectation values of the exciton operators, is a consequence of the linearization of quantum fluctuations. However the present approach includes the light field quantization and can thus be applied to the description of quantum optical phenomena. Indeed, striking difference between the semiclassical and the full quantum descriptions emerge when considering expectation values of exciton and photon numbers or even higher order correlators. For this reason, in the following section we derive operatorial dynamical equations useful for the calculation of such higher order correlators. By explicit calculation it is easy to see that the first term in Eq. (4.22) is zero unless all the involved polarization labels $\sigma$ coincide. In order to manipulate the last term we follow the procedure of Ref. [24] which succeeded in reformulating the nonlinear term coming from the Coulomb interaction as an exciton-exciton (X-X) mean-field contribution plus a correlation term driven by a two-exciton correlation function. Even if we are about to perform more or less the same steps of Ref. [24] we shall provide a detailed account of all the key points of the present derivation. A clear comprehension of these details will be essential for the extension to operatorial dynamical equations of the next section.

In order to manipulate the last term we shall need the two identities:

$$c_{nk;\tilde{n}k;\tilde{n}k}^{(1)} = (\tilde{\omega}x - \tilde{\omega}_{1\tilde{n}k} - \tilde{\omega}_{1nk}) \langle 1\tilde{n}\tilde{k} | B_{nk} | 2\beta \rangle =$$

$$= \langle 1\tilde{n}\tilde{k} | B_{nk}(\tilde{H}_c - \tilde{\omega}_{1\tilde{n}k} - \tilde{\omega}_{1nk}) | 2\beta \rangle$$

(4.24)
\[
\frac{d}{dt} \left( \langle \hat{B}_{n'k'}(1) | \langle \hat{B}_{n''k''}(1) e^{-i\Omega(t-t')} \right) = \]
\[
= +i \left( V_{n'k'} \langle \hat{a}_{k'}(1) \langle \hat{B}_{n''k''}(1) + V_{n'k'} \langle \hat{a}_{k'}(1) \langle \hat{B}_{n''k''}(1) e^{-i\Omega(t-t')} \right),
\]

or
\[
\frac{1}{2} \frac{d}{dt} \sum_{n'k' : n''k''} \left( \langle \hat{B}_{n'k'}(1) \langle \hat{B}_{n''k''}(1) e^{-i\Omega(t-t')} \right) = +i \sum_{n'k' : n''k''} V_{n'k'} \langle \hat{a}_{k'}(1) \langle \hat{B}_{n''k''}(1) e^{-i\Omega(t-t')},
\]

where \( \Omega = \omega_{1n'k'} - \omega_{1n''k''} - 2i \Gamma_x \). We have:

\[
\sum_{\beta} \langle \hat{X}_{0;\beta} | 2\beta \rangle = \sum_{\beta} \langle 1\hat{n}\tilde{k} | \hat{B}_{nk}(\hat{H}_e - \tilde{\omega}_{1\hat{n}\tilde{k}} - \omega_{1nk}) | 2\beta \rangle \cdot \]

\[
i \sum_{n'k' : n''k''} V_{n'k'} \langle 2\beta | \hat{B}_{n'k'}^{\dagger} | 1n''k'' \rangle \int_{-\infty}^{t} dt' e^{-i(\omega_{2\beta} - i\Gamma_{xx})(t-t')} \langle \hat{a}_{k'}(1)(t') \langle \hat{B}_{n''k''}(1)(t') \right).
\]

We observe that the matrix elements entering the nonlinear source terms are largely independent on the wave vectors for the range of wave vectors of interest in the optical response. Neglecting such dependence we can thus exploit the identity (4.26), obtaining

\[
\hbar \sum_{\beta} \langle 1\hat{n}\tilde{k} | \hat{B}_{nk}(\frac{\hat{H}_e}{\hbar} - \omega_{1\hat{n}\tilde{k}} - \omega_{1nk}) | 2\beta \rangle \int_{-\infty}^{t} dt' e^{-i(\omega_{2\beta} - i\Gamma_{xx})(t-t')} \]

\[
= \hbar \sum_{n'k' : n''k''} \langle 2\beta | \hat{B}_{n'k'}^{\dagger} | 1n''k'' \rangle \frac{1}{2} \frac{d}{dt'} \left( \langle \hat{B}_{n'k'}(1)(t') \langle \hat{B}_{n''k''}(1)(t') e^{-i\Omega(t-t')} \right) e^{+i\Omega(t-t')} = \]

\[
= \hbar \sum_{n'k' : n''k''} \int_{-\infty}^{t} dt' \langle 1\hat{n}\tilde{k} | \hat{B}_{nk}(\frac{\hat{H}_e}{\hbar} - \omega_{1\hat{n}\tilde{k}} - \omega_{1nk}) e^{-i\tilde{\omega}_{1\hat{n}\tilde{k}}(t-t')} \hat{B}_{n'k'}^{\dagger} | 1n''k'' \rangle e^{-\Gamma_{xx}(t-t')} \]

\[
\frac{1}{2} \frac{d}{dt'} \left( \langle \hat{B}_{n'k'}(1)(t') \langle \hat{B}_{n''k''}(1)(t') e^{-i\Omega(t-t')} \right) e^{+i\Omega(t-t')},
\]

\[
(4.28)
\]
where in the last lines we have resummed all the biexciton subspace by virtue of its completeness. We shall perform an integration by part.

\[
\begin{align*}
\frac{1}{2\hbar} \sum_{n'k',n''k''} \left\{ e^{i(\omega_{1n'k'} + \omega_{1n''k''} - 2i\Gamma_x + i\Gamma_{xx})(t-t')} \langle 1\tilde{n}\tilde{k} | \hat{B}_{nk} \left( \frac{\hat{H}_c}{\hbar} - \omega_{1\tilde{n}\tilde{k}} - \omega_{1nk} \right) e^{-i(\frac{\Omega}{2}(t-t') - \frac{\Omega}{2}(t'))} \hat{B}_{n'k'}^\dagger | 1n''k'' \rangle \right\} & \\
& - \int_{-\infty}^{t} dt' \langle 1\tilde{n}\tilde{k} | \hat{B}_{nk} \left( \frac{\hat{H}_c}{\hbar} - \omega_{1\tilde{n}\tilde{k}} - \omega_{1nk} \right) e^{-i(\frac{\Omega}{2}(t-t') - \frac{\Omega}{2}(t'))} \hat{B}_{n'k'}^\dagger | 1n''k'' \rangle \right\} \\
& = \frac{1}{2\hbar} \sum_{n'k',n''k''} \left\{ \langle 1\tilde{n}\tilde{k} | \hat{B}_{nk} \left( \frac{\hat{H}_c}{\hbar} - \omega_{1\tilde{n}\tilde{k}} - \omega_{1nk} \right) \hat{B}_{n'k'}^\dagger | 1n''k'' \rangle \langle \hat{B}_{n'k'} \rangle (1) \langle \hat{B}_{n''k''} \rangle (1) \right\} \right. \\
& \quad - \int_{-\infty}^{t} dt' \langle 1\tilde{n}\tilde{k} | \hat{B}_{nk} \left( \frac{\hat{H}_c}{\hbar} - \omega_{1\tilde{n}\tilde{k}} - \omega_{1nk} \right) \hat{B}_{n'k'}^\dagger | 1n''k'' \rangle \right\} \\
& = \frac{1}{2\hbar} \sum_{n'k',n''k''} \left\{ \langle 1\tilde{n}\tilde{k} | \hat{B}_{nk} \left( \frac{\hat{H}_c}{\hbar} - \omega_{1\tilde{n}\tilde{k}} - \omega_{1nk} \right) \hat{B}_{n'k'}^\dagger | 1n''k'' \rangle \langle \hat{B}_{n'k'} \rangle (1) \langle \hat{B}_{n''k''} \rangle (1) \right\} \\
& \quad - \int_{-\infty}^{t} dt' \langle 1\tilde{n}\tilde{k} | \hat{B}_{nk} \left( \frac{\hat{H}_c}{\hbar} - \omega_{1\tilde{n}\tilde{k}} - \omega_{1nk} \right) \hat{B}_{n'k'}^\dagger | 1n''k'' \rangle \right\}.
\end{align*}
\]

The first and the second term can be expressed in terms of a double commutator structure:

\[
\langle 1\tilde{n}\tilde{k} | \hat{B}_{nk} \left( \frac{\hat{H}_c}{\hbar} - \omega_{1\tilde{n}\tilde{k}} - \omega_{1nk} \right) = \langle 0 | [\hat{B}_{n\tilde{k}}, [\hat{B}_{nk}, \hat{H}_c]] \approx \langle 0 | \hat{D}_{n\tilde{k},nk} \rangle.
\]
where a force operator $\hat{D}$ is defined [24] and

$$\begin{align*}
\frac{d}{dt'} \left\{ e^{i(\omega_{1n',k'} + \omega_{1n,k'} - 2\Gamma_x + i\Gamma_x)(t-t')} \langle \hat{\tau}_{n, n'} \rangle \right\} = \\
\langle 1\tilde{n}, \tilde{k} | \hat{B}_{nk}(\hat{H}_c - \omega_{1\tilde{n}, \tilde{k}} - \omega_{1nk}) e^{-i\hat{H}_c/\hbar (t-t')} \hat{B}_{n'k'}^\dagger | 1n'n'' \rangle = \\
\frac{d}{dt'} \left\{ \langle 0 | \hat{D}_{\tilde{n}, n', n''} e^{-i\hat{H}_c/\hbar (t-t')} \hat{B}_{n'k'}^\dagger \hat{B}_{n''k''}^\dagger | 0 \rangle e^{i(\omega_{1n', k'} + \omega_{1n', k''} - 2i\Gamma_x + i\Gamma_x)(t-t')} \right\} = \\
= \langle 0 | \hat{D}_{\tilde{n}, n', n''} e^{-i\hat{H}_c/\hbar (t-t')} i\left( \hat{H}_c - \omega_{1n', k'} - \omega_{1n', k''} - i(\Gamma_{xx} - 2\Gamma_x) \right) \\
\hat{B}_{n'k'}^\dagger \hat{B}_{n''k''}^\dagger | 0 \rangle e^{i(\omega_{1n', k'} + \omega_{1n', k''} - 2i\Gamma_x + i\Gamma_x)(t-t')} = \\
e^{i(\omega_{1n', k'} + \omega_{1n', k''} - 2i\Gamma_x + i\Gamma_x)(t-t')} i\Gamma_{nk, nk'} \hat{B}_{n'k'}^\dagger \hat{B}_{n''k''}^\dagger (t - t') + \\
(\Gamma_{xx} - 2\Gamma_x) e^{i(\omega_{1n', k'} + \omega_{1n', k''} - 2i\Gamma_x + i\Gamma_x)(t-t')} \langle 0 | \hat{D}_{\tilde{n}, n', nk} (t - t') \hat{B}_{n'k'}^\dagger \hat{B}_{n''k''}^\dagger | 0 \rangle ,
\end{align*}$$

(4.32)

where the memory kernel reads

$$\Gamma_{nk, nk'}^{n, n''} (t - t') = \langle 0 | \hat{D}_{nk, nk'} (t - t') \hat{D}_{n''k''}^\dagger | 0 \rangle .$$

(4.33)

The usual time dependence in the Heisenberg picture is given by $\hat{D}(t) = e^{i\hat{H}_c\tau} \hat{D} e^{-i\hat{H}_c\tau}$.

Altogether, the nonlinear term originating from Coulomb interaction can be written as

$$Q_{nk, \tilde{n}k}^{\text{COUL}(2)} = \sum_{\beta} e_{nk, \tilde{n}k; \beta}^{(1)} (\hat{X}_{0; 23})^{(2)} =$$

$$\begin{align*}
\frac{1}{2} \hbar \sum_{n', n''} \left\{ \langle 0 | \hat{D}_{nk, n'} \hat{B}_{n'k'}^\dagger \hat{B}_{n''k''}^\dagger | 0 \rangle \langle \hat{B}_{n'k'} \rangle^{(1)} (t) \langle \hat{B}_{n''k''} \rangle^{(1)} (t) - \\
-i \int_{-\infty}^t dt'' \Gamma_{nk, nk'}^{n', n''} (t - t'') \langle \hat{B}_{n'k'} \rangle^{(1)} (t') \langle \hat{B}_{n''k''} \rangle^{(1)} (t') e^{-\Gamma_{xx}(t-t'')} \right\} - \\
- \frac{\hbar}{2} (\Gamma_{xx} - 2\Gamma_x) \sum_{n', n''} \int_{-\infty}^t dt'' \langle 0 | \hat{D}_{nk, nk'} (t - t'') \hat{B}_{n'k'}^\dagger \hat{B}_{n''k''}^\dagger | 0 \rangle \langle \hat{B}_{n'k'} \rangle^{(1)} (t') \langle \hat{B}_{n''k''} \rangle^{(1)} (t') .
\end{align*}$$

(4.34)

For later purpose we are interested in the optical response dominate by the 1S exciton sector, with $\Gamma_{xx} \simeq 2\Gamma_x$ in the cases of counter- and co-circularly polarized
waves. Specifying to this case the Coulomb-induced term with Eq. (4.34) becomes

\[
\frac{d}{dt} \langle \hat{\mathbf{B}}_{\pm \mathbf{k}} \rangle^{(3)}_{\text{COUL}} = -i\tilde{\omega}_k \langle \hat{\mathbf{B}}_{\pm \mathbf{k}} \rangle^{(3)} - \frac{i}{\hbar} \sum_{\tilde{\sigma} \mathbf{k}} \langle \hat{\mathbf{B}}_{\tilde{\sigma} \mathbf{k}} \rangle^{(1)}(t) \langle \hat{\mathbf{B}}_{\pm \mathbf{k}} \rangle^{(1)}(t) \langle \hat{\mathbf{B}}_{\pm \mathbf{k}} \rangle^{(1)}(t) +
\]

\[
- \frac{1}{\hbar} \sum_{\sigma' \sigma'' \tilde{\sigma} \mathbf{k}' \mathbf{k}''} \delta_{\mathbf{k} + \mathbf{k}' + \mathbf{k}''} \delta_{\pm + \tilde{\sigma} \sigma' + \sigma''} \langle \hat{\mathbf{B}}_{\tilde{\sigma} \mathbf{k}} \rangle^{(1)}(t)
\]

\[
\int_{-\infty}^{t} dt' F^{\sigma' \sigma''}(t - t') \langle \hat{\mathbf{B}}_{\sigma' \mathbf{k}'} \rangle^{(1)}(t') \langle \hat{\mathbf{B}}_{\sigma'' \mathbf{k}''} \rangle^{(1)}(t') e^{-\Gamma_{xx}(t-t')},
\]

where, in order to lighten the notation, we dropped the two spin indexes \(\sigma\) and \(\tilde{\sigma}\) in the four-particle kernel function \(F\) defined in Eq. (4.33) for they are already univocally determined once chosen the others (i.e. \(\sigma'\) and \(\sigma''\)) as soon as their selection rule \((\delta_{\sigma + \tilde{\sigma} \sigma' + \sigma''})\) is applied. Moreover, the \(\hbar/2\) has been reabsorbed in the Coulomb nonlinear coefficients \(V_{xx}\) and \(F^{\sigma' \sigma''}(t - t')\). A detail microscopic account for the mean-field \(V_{xx}\), for the \(F\)’s and their selection rules are considered in [69]. For the range of \(k\)-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, \(F^{\pm +}(t - t')\) and \(F^{+ \pm}(t - t')\) (counter-circular polarization) give rise to a mixing between the two circularly polarizations. The physical origin of the three terms in Eq. (4.34) 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 [24]. Equation (4.34) even if formally similar to that of Ref. [24], represents its extension including polaritonic effects due to the presence of the cavity. It has been possible thanks to the inclusion of the dynamics of the cavity modes whereas in Ref. [24]
the electromagnetic field entered as a parameter only. Former analogous extensions have been obtained within a semiclassical model [69, 66]. 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. [66], 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 respect to that of excitons in bare QWs, thus producing a modification of the integral in Eq. (4.34). In this way the exciton-photon coupling $V_{nk}$ affects the exciton-exciton collisions that govern the polariton amplification process. Ref. [66] considers the

![Figure 4.1](image)

Figure 4.1. Figure 1 of Ref. [66]. Energy dependence of the effective exciton-exciton scattering potential $\mathcal{F}(\omega)$, calculated for a GaAs QW 7 nm wide with an exciton binding energy of 13.5 meV using a T-matrix approach.

first (mean-field) and the second (four-particle correlation) terms in the particular case of cocircularly polarized waves, calling them without indexes as $V_{xx}$ and $F(t)$ respectively. In Fig. 4.1 they show $\mathcal{F}(\omega)$, the Fourier transform of $F(t)$ plus the
mean-field term $V_{xx}$,

$$
\mathcal{F}(\omega) = V_{xx} - i \int_{-\infty}^{\infty} dt F(t) e^{i\omega t}.
$$  \hspace{1cm} (4.36)

Its imaginary part is responsible for the frequency dependent excitation induced dephasing, it reflects the density of the states of two-exciton pair coherences. Towards the 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 [66] that both effects depends on the sum of the energies of the scattered polariton pairs. The third term in Eq. (4.34) can be thought as a reminder of the mismatch between the picture of a biexciton as a composite pair of exciton. In the following we will set $\Gamma_{xx} \simeq 2\Gamma_x$.

The other nonlinear source term in Eq. (4.21) depends directly on the exciton wave function and reads

$$
\sum_{\tilde{n}\k} (\hat{B}_{\tilde{n}\k})^*(1) \sum_{n'k',n''k''} C_{\tilde{n}\k,nk}^{n'k',n''k''} \langle \hat{a}_{k'}^{(1)} \rangle \langle \hat{B}_{n''k''}^{(1)} \rangle.
$$  \hspace{1cm} (4.37)

It represents a phase-space filling (PSF) contribution, due to the Pauli blocking of
electrons. It can be developed as follows,

\[
C_{\tilde{n}\tilde{k},nk}^{n'k',n''k''} = V_{n'k'} \langle 1\tilde{n}\tilde{\sigma}\tilde{k} | \delta_{(n'k'):(nk)} - [\hat{B}_{n\sigma k},\hat{B}_{n'\sigma' k'}^\dagger] | 1n''\sigma''k'' \rangle =
\]

\[
= V_{n'k'} \delta_{\sigma,\sigma'} \left\{ \sum_q \Phi_{n'\sigma q}^k \Phi_{n''\sigma' q'}^{k'} \langle 1\tilde{n}\tilde{\sigma}\tilde{k} | \vec{c}_{\sigma,q}^\dagger \delta_{\sigma',q + \eta_h (k'' - k') + \eta_r k + \eta_r k} | 1n''\sigma''k'' \rangle + \right.
\]

\[
\sum_q \Phi_{n'\sigma q}^k \Phi_{n''\sigma' q'}^{k'} \langle 1\tilde{n}\tilde{\sigma}\tilde{k} | \delta_{\sigma',q + \eta_h (k'' - k') + \eta_r k} \rangle \delta_{\sigma,\sigma'} d_{\sigma',q + \eta_h k} | 1n''\sigma''k'' \rangle \left. \right\} =
\]

\[
= V_{n'k'} \delta_{\sigma,\sigma'} \delta_{\tilde{k} + \tilde{k}',\tilde{k}''} \left\{ \sum_q \Phi_{n'\sigma q}^k \Phi_{n''\sigma' q'}^{k'} \Phi_{n''\sigma'' q''}^{k''} + \sum_q \Phi_{n'\sigma q}^k \Phi_{n''\sigma' q'}^{k'} \Phi_{n''\sigma'' q''}^{k''} \right\},
\]

the explicit expressions of the \( q \)'s are such that

\[
q_0 = q + \eta_h (k' - k), \quad q_3 = q - \eta_r (k' - k)
\]

\[
q_1 = q - k'' + \eta_h \tilde{k} + \eta_r k, \quad q_4 = q + k'' - \eta_h \tilde{k} - \eta_r k
\]

\[
q_2 = q - k'' + \eta_h k + \eta_r k'', \quad q_5 = q + k'' - \eta_h k - \eta_r k''.
\]

Thus, the nonlinear dynamics of Eq. (4.20) driven by \( \hat{H}_I \) can be written

\[
\frac{d}{dt} \langle \hat{B}_{n\sigma k} \rangle^{(3)} \bigg|_{\hat{H}_I} = +i \frac{V_{n\sigma k}}{\hbar} \langle \hat{a}_{\sigma k} \rangle^{(3)} - i \frac{\hbar}{V_{n'\sigma q}} \sum_{k''} \delta_{\tilde{k} + \tilde{k}',\tilde{k}''} \langle \hat{B}_{n'\sigma q} \rangle^{(1)}
\]

\[
\langle \hat{a}_{\sigma k'} \rangle^{(1)} \langle \hat{B}_{n''\sigma' k''} \rangle^{(1)} \langle \hat{c}_{\sigma',q}^\dagger \delta_{\sigma',q + \eta_h (k'' - k') + \eta_r k + \eta_r k} | 1n''\sigma''k'' \rangle \sum_q \Phi_{n'\sigma q}^k \Phi_{n''\sigma' q'}^{k'} \Phi_{n''\sigma'' q''}^{k''} + \sum_q \Phi_{n'\sigma q}^k \Phi_{n''\sigma' q'}^{k'} \Phi_{n''\sigma'' q''}^{k''}\bigg|_{\hat{H}_I}^{(3)}
\]

We are interested in studying polaritonic effects in SMCs where the optical response involves mainly excitons belonging to the 1S band with wave vectors close to normal incidence, i.e. \(|k| \ll \frac{\pi}{a_x}\) (much lower than the inverse of the exciton Bohr radius).

In this case the exciton relative wave functions are independent on spins as well as on the center of mass wave vector. They are such that \( \sum_{q=-\infty}^{\infty} |\Phi_q|^2 = 1 \), i.e.

\[
\Phi_q = \frac{1}{\sqrt{A}} \frac{\sqrt{2} a_x}{(1+(a_x|q|)^2)^{1/2}}, \quad a_x \text{ is the exciton Bohr radius. From now on whenever no}
\]

\[71\]
excitonic level is specified the 1S label is understood. It yields
\[
\frac{d}{dt} \langle \hat{B}_{\sigma k} \rangle^{+}(3) \big|_{\hat{H}_I} = +i \frac{V_{\sigma k}}{\hbar} \langle \hat{a}_{\sigma k} \rangle^{+}(3) - \frac{i}{\hbar} \sum_{k'k''} \delta_{k+k',k''} \langle \hat{B}_{\sigma k''} \rangle^{*}(1) \langle \hat{a}_{\sigma k'} \rangle^{(1)} \langle \hat{B}_{\sigma k} \rangle^{(1)} 2\hat{V}_\sigma O_{PSF}^{PSF},
\]  
(4.40)
where the overlap \(O_{PSF}^{PSF}\) has been calculated in the case of zero center of mass wave vector, namely
\[
O_{PSF} = \sum_\Phi \Phi^* \Phi \Phi^* \Phi.
\]
In SMCs a measured parameter is the so-called vacuum Rabi splitting \(V_{\sigma k}\) [12] of the 1S excitonic resonance, for the range of \(k\)-space of interest essentially constant. Defining \(V = \hat{V}_\sigma \sqrt{A\phi^*(0)}\)
\[
V\sigma O_{PSF} = \frac{V}{\sqrt{A\phi^*(0)}} O_{PSF} = \frac{8\pi a_s^2}{7A} V = \frac{1}{2} \frac{V}{n_{sat}},
\]  
(4.41)
where we have set \(n_{sat} = (7/16)\langle A/\pi a_s^2 \rangle\), called saturation density.

In terms of the two circular polarizations the dynamics induced by \(\hat{H}_I\) finally reads
\[
\frac{d}{dt} \langle \hat{B}_{\pm k} \rangle^{+}(3) \big|_{\hat{H}_I} = +i \frac{V}{\hbar} \langle \hat{a}_{\pm k} \rangle^{+}(3) - \frac{i}{\hbar} \sum_{k} \langle \hat{B}_{\pm k} \rangle^{*}(1) Q_{PSF(2)}^{PSF},
\]  
(4.42)
where
\[
\sum_{k} \langle \hat{B}_{\pm k} \rangle^{*}(1) Q_{PSF(2)}^{PSF} = \frac{V}{n_{sat}} \sum_{k'k''} \delta_{k+k',k''} \langle \hat{B}_{\pm k''} \rangle^{*}(1) \langle \hat{a}_{\pm k'} \rangle^{(1)} \langle \hat{B}_{\pm k} \rangle^{(1)}. 
\]  
(4.43)
The same lines of argument can be followed for computing the Coulomb-induced interactions \(Q_{COUL}^{COUL(2)}\) [69].

We are lead to introduce the saturation density for two main reasons. The most obvious is our interest to refer this work to the literature where \(n_{sat}\) is extensively used [14, 21, 66, 67]. The other most interesting reason is that we can directly
compute this quantity. Indeed, the equation of motion for the exciton operator reads

$$\frac{d}{dt} \langle \hat{B}_{\pm k} \rangle^{(3)} = -i\tilde{\omega}_k \langle \hat{B}_{\pm k} \rangle^{(3)} + i \frac{V}{\hbar} \langle \hat{a}_{\pm k} \rangle^{(3)} - i \frac{V}{\hbar} \sum_{\tilde{\sigma} = \pm k} \langle \hat{B}_{\tilde{\sigma} k} \rangle^{(1)} Q^{COUL(2)}_{\pm k, \tilde{\sigma} k}$$

$$- i \frac{2}{\hbar} \frac{V}{\sqrt{A\phi^* (0)}} O^{PSF} \sum_{k' k''} \delta_{k' k''} \langle \hat{B}_{\pm \tilde{k}} \rangle^{(1)} \langle \hat{a}_{\pm k'} \rangle^{(1)} \langle \hat{B}_{\pm k''} \rangle^{(1)}.$$  

Leaving apart the discrepancy between the order in the DCTS we can compute the so-called oscillator strength (OS), defined as what multiplies the photon expectation values $\langle \hat{a}_{\pm k=0} \rangle$,

$$OS = i \frac{V}{\hbar} \left( 1 - \frac{2}{\sqrt{A\phi^* (0)}} O^{PSF} \left[ \langle \hat{B}_{\pm 0} \rangle^{(1)} \langle \hat{B}_{\pm 0} \rangle^{(1)} \right] \right).$$  

(4.44)

The saturation density may be defined as the exciton density that makes the oscillator strength to be zero. We obtain

$$n_{sat} = \left( \frac{2}{\sqrt{A\phi^* (0)}} O^{PSF} \right)^{-1} = \frac{A}{\pi a_x^2} \frac{7}{16}. \quad (4.45)$$

Eventually, the lowest order ($\chi^{(3)}$) nonlinear optical response in SMCs are described by the following set of coupled equations:

$$\frac{d}{dt} \langle \hat{a}_{\pm k} \rangle^{(3)} = -i\tilde{\omega}_k \langle \hat{a}_{\pm k} \rangle^{(3)} + i \frac{V}{\hbar} \langle \hat{B}_{\pm k} \rangle^{(3)} + t_c \frac{E_{\pm k}}{\hbar}, \quad (4.46)$$

$$\frac{d}{dt} \langle \hat{B}_{\pm k} \rangle^{(3)} = -i\tilde{\omega}_k \langle \hat{B}_{\pm k} \rangle^{(3)} + i \frac{V}{\hbar} \langle \hat{a}_{\pm k} \rangle^{(3)} - i \frac{V}{\hbar} \sum_{\tilde{\sigma} = \pm k} \langle \hat{B}_{\tilde{\sigma} k} \rangle^{(1)} R^{(2)}_{\pm k, \tilde{\sigma} k}, \quad (4.47)$$

with $\sum_{\tilde{\sigma} k} \langle \hat{B}_{\tilde{\sigma} k} \rangle^{(1)} R^{(2)}_{\pm k, \tilde{\sigma} k} = \sum_{\tilde{\sigma} k} \langle \hat{B}_{\tilde{\sigma} k} \rangle^{(1)} Q^{COUL(2)}_{\pm k, \tilde{\sigma} k} + \sum_{\tilde{k}} \langle \hat{B}_{\pm \tilde{k}} \rangle^{(1)} Q^{PSF(2)}_{\pm \tilde{k}, \tilde{\sigma} \tilde{k}}$, with the first of the two addenda originating from Coulomb interaction, Eq. (4.35), whereas the second represents the phase-space filling contribution written in Eqs. (4.43). Starting from here, in the strong coupling case, it might be useful to transform the description into a polariton basis. The proper inclusion of dephasing/relaxation and
the application of these equations to parametric processes, in the strong coupling regime, is described in another chapter [25].

Equations (4.46) and (4.47) is exact to the third order in the exciting field. While a systematic treatment of higher-order optical nonlinearities would require an extension of the equations of motions (see e.g. Appendix), a restricted class of higher-order effects can be obtained from solving equations (4.46) and (4.47) self-consistently up to arbitrary order as it is usually employed in standard nonlinear optics. This can be simply accomplished by replacing, in the nonlinear sources, the linear excitonic polarization and light fields with the total fields [24, 66, 70]. Multiple-scattering processes are expected to be very effective in cavity-embedded QWs due to multiple reflections at the Bragg mirrors.

4.2 Coherent optical parametric processes for polaritons

We start from the Heisenberg equations of motion for the exciton and photon operators. In the DCTS spirit, we keep only those terms providing the lowest nonlinear response ($\chi^{(3)}$) in the input light field [71]. We assume the pump polaritons driven by a quite strong coherent input field $E_k^m = \langle \hat{E}_{ik}^{(+)} \rangle$ consisting of a classical (C-number) field, resonantly exciting the structure at a given energy and wave vector, $k_p$. We are interested in studying polaritonic effects in SMCs where the optical response involves mainly excitons belonging to the 1S band with wave vectors close to normal incidence, $|k| \ll \frac{\pi}{a_x}$. We retain only those terms containing the semiclassical pump amplitude twice, thus focusing on the “direct” pump-induced nonlinear parametric interaction. One ends up with a set of coupled equations of motion exact to the third order in the exciting field. While a systematic treatment of higher-order
optical nonlinearities would require an extension of the equations of motion, a
restricted class of higher-order effects can be obtained from solving these equations
self-consistently up to arbitrary order as it is usually employed in standard nonlin-
ear optics. This can be simply accomplished by replacing, in the nonlinear sources,
the linear excitonic polarization and and light field operators with the total field.
From now on, since the pump-driven terms (e.g. the $B$ and $a$ at $k_p$) are $\mathbb{C}$-numbers
coherent amplitudes like the semiclassical electromagnetic pump field, we will make
such distinction in marking with a “hat” the operators only. It yields [71]

$$\dot{\hat{B}}_k = -i\omega_k \hat{B}_k - i\delta_k + iV\hat{a}_k - i\hat{R}^{NL}_k, \quad (4.48a)$$

$$\dot{\hat{a}}_k = -i\omega_k \hat{a}_k + iV\hat{B}_k + t_c E_{in}^{in}; \quad (4.48b)$$

where $\omega_k(i = x, c)$ are the energies of QWs excitons and cavity photons. The
intracavity and the exciton field of a given mode $k$ are coupled by the exciton-cavity
photon coupling rate $V$. The relevant non-linear source term, able to couple waves
with different in-plane wave vector $k$, is given by

$$\dot{\hat{R}}^{NL}_k = (\hat{R}^{sat}_k + \hat{R}^{xx}_k)/N_{eff};$$

where the first term originates from the phase-space filling of the exciton transition,

$$\hat{R}^{sat}_k = V n_{sat} \hat{B}_k \hat{a}_k \hat{B}_k^\dagger; \quad (4.49)$$

being $n_{sat} = 7/16$ the exciton saturation density and $k_i = 2k_p - k$. $N_{eff}$ depends
on the number of wells inside the cavity and their spatial overlap with the cavity-
mode. Inserting a large number of QWs into the cavity results also in increasing
the photon-exciton coupling rate $V = V_1 \sqrt{N_{eff}}$, where $V_1$ is the exciton-photon
coupling for 1 QW. $\hat{R}^{xx}_k$ is the Coulomb interaction term. It dominates the coherent
xx coupling and for co-circularly polarized waves (the only case here addressed) can
be written as

$$\dot{\hat{R}}^{xx}_k = \hat{B}^{\dagger}_{k_i}(t) \left( V_{xx} B_{k_p}(t) B_{k_p}(t) - \right.$$

$$\left. -i \int_{-\infty}^t dt' F(t - t') B_{k_p}(t') B_{k_p}(t') \right), \quad (4.50)$$

75
where $V_{xx} \simeq 6E_b/\pi$, being $E_b$ the exciton binding energy. Equation (4.50) includes the instantaneous mean-field $xx$ interaction term and a non-instantaneous term originating from four-particle correlations. These equations show a close analogy to those derived in [47], addressing the bulk case. In addition to that former result, in the present formulation we succeed in dividing rigorously (in the DCTS spirit) the Coulomb-induced correlations into mean-field and four-particle correlation terms. Moreover the pump-induced shift due to parametric scattering $\hat{s}_k$ reads

$$N_{eff}\hat{s}_k = \frac{V}{n_{sat}} \left( B^*_{k_p}a_{k_p}\hat{B}_k + B^*_{k_p}B_{k_p}\hat{a}_k \right) + 2V_{xx}B^*_{k_p}B_{k_p}\hat{B}_k - 2iB^*_{k_p}(t) \int_{-\infty}^{t} dt' F(t-t')\hat{B}_k(t')B_{k_p}(t').$$

Equation (4.48) can be written in compact form as

$$\dot{B}_k = -i\Omega_{kk}^{xc}B_k + \mathcal{E}^\text{in}_k - i\mathcal{R}_{kk}^{NL};$$

where $B_k \equiv \left( \hat{B}_k \atop \hat{a}_k \right)$, $\Omega_{kk}^{xc} \equiv \left( \begin{array}{cc} \omega^x_k & -V \\ -V & \omega^c_k \end{array} \right)$, $\mathcal{E}_{k}^{\text{in}} \equiv \left( \begin{array}{c} 0 \\ t_cE_k^{\text{in}} \end{array} \right)$, and $\mathcal{R}_{kk}^{NL} \equiv \left( \begin{array}{c} \hat{s}_k + \hat{R}_{kk}^{NL} \\ 0 \end{array} \right)$. 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 $\Omega_{kk}^{xc}$. The coupling rate $V$ determines the splitting ($\simeq 2V$) between the two polariton energy bands. This nonperturbative dynamics including the interactions (induced by $\hat{R}_{kk}^{NL}$) between different polariton modes can be accurately described by Eq. (4.48). Nevertheless there can be reasons to prefer a change of bases 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. In order to obtain the dynamics for the polariton system we perform on the exciton and photon operators the unitary basis transformation

$$P_k = U_k B_k;$$

being $$P_k = \begin{pmatrix} \hat{P}_{1k} \\ \hat{P}_{2k} \end{pmatrix}$$ and

$$U_k = \begin{pmatrix} X_{1k} & C_{1k} \\ X_{2k} & C_{2k} \end{pmatrix}. $$

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

$$[\hat{B}_n, \hat{B}^\dagger_{n'}] = \sum_{q} \Phi_{nq}^* \Phi_{n'q} \sum_{\alpha, \beta} \left( \langle N\alpha | \hat{c}_{q}^\dagger \hat{c}_{q} | N\beta \rangle + \langle N\alpha | \hat{d}_{q}^\dagger \hat{d}_{-q} | N\beta \rangle \right) | N\alpha \rangle \langle N\beta |. $$

Thus, within a DCTS line of reasoning [24], 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, they are density-dependent contributions. Evidently all these considerations affect polariton statistics as well, being polariton 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 they obey approximately bosonic-like commutation rules.

Diagonalizing $\Omega_k^{xc}$:

$$U_k \Omega_k^{xc} = \tilde{\Omega}_k U_k,$$

where

$$\tilde{\Omega}_k = \begin{pmatrix} \omega_{1k} & 0 \\ 0 & \omega_{2k} \end{pmatrix}.$$  

$\omega_{1,2}$ are the eigenenergy (as a function of $k$) of the lower (1) and upper (2) polariton states. After simple algebra it is possible to obtain this relation for the Hopfield coefficients [21]:

$$X_{1k} = -C_{2k}^*; \quad C_{1k} = X_{2k}^*.$$  

where

$$X_{1k} = \frac{1}{\sqrt{1 + \left(\frac{V}{\omega_{1k} - \omega_c}ight)^2}} \quad C_{1k} = \frac{1}{\sqrt{1 + \left(\frac{\omega_{1k} - \omega_c}{V}ight)^2}}.$$  

Introducing this transformation into Eq. (4.52), one obtains

$$\dot{P}_k = -i\tilde{\Omega}_k P_k + \tilde{E}_{in}^k - i\tilde{R}_{NL}^k;$$  

where $\tilde{R}_{NL}^k = U R_{NL}^k$, which in explicit form reads

$$\dot{P}_{1k} = -i\omega_{1k} P_{1k} - i\tilde{s}_{1k} + \tilde{E}_{1k}^m - i\tilde{R}_{1k}^m;$$  

$$\dot{P}_{2k} = -i\omega_{2k} P_{2k} - i\tilde{s}_{2k} + \tilde{E}_{2k}^m - i\tilde{R}_{2k}^m;$$

where $\tilde{E}_{nm}^m = t_c C_{mk} E_{nk}^{in}$, and $\tilde{R}_{mk} = X_{mk} R_{k}^{NL}$, $(m = 1,2)$. Such a diagonalization is the necessary step when the eigenstates of the polariton system are to be used as the starting states perturbed by the interaction with the environment degrees of freedom [79]. The nonlinear interaction written in terms of polariton operators reads

$$\tilde{R}_{NL}^k = \sum_{i,j,l} \tilde{P}_{ik}^\dagger (t) \int_{-\infty}^t g_{ijl}^{kl}(t,t') P_{jk}(t') P_{lk}(t') dt',$$
being

\[ g_{mk}^{ijl}(t,t') = \frac{1}{N_{\text{eff}}} \left[ \frac{V}{n_{\text{sat}}} C_{j,kp}^* \delta(t-t') + \left( V_{xx} \delta(t-t') - iF(t-t') \right) X_{j,kp}^* \right] X_{l,kp}^* X_{i,k}^* . \] (4.62)

The shift \( \tilde{s}_k(t) \) is transformed into

\[ \tilde{s}_{mk}(t) = \sum_{i,j,l} P_{ikp}^* (t) \int_{-\infty}^t \left( h_{mk}^{ijl} \delta(t-t') - 2iF(t-t') \right) P_{jkp}(t') \tilde{P}_{lk}(t') dt' , \] (4.63)

and

\[ h_{mk}^{ijl} = \frac{1}{N_{\text{eff}}} X_{mk} \left[ \frac{V}{n_{\text{sat}}} X_{ikp} \left( C_{j,kp}^* X_{lk}^* + X_{jkp}^* C_{lk}^* \right) + 2V_{xx} X_{ikp}^* X_{jkp}^* X_{lk}^* \right] . \] (4.64)

Equation (4.60) 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. Of course there are nonlinear optical processes involving modes of only one branch [19, 17]. In this case it is possible to take into account only one of the two set of equations in (4.60) and to eliminate the summation over the branch indexes in Eq. (4.61).

Analogous equations can be obtained starting from an effective Hamiltonian describing excitons as interacting bosons [21]. 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. (4.48) have nonlinear terms of the same structure of Ref. [21] (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

\[
R_{NL}^k \big|_{psf} = \frac{V}{n_{sat}} \sum_{k',q} C_{k' - q} X_{k+q} X_{k'} P_{k'}^* P_{k+q} P_{k'-q} .
\] (4.65)

the corresponding term in [21] can be written as

\[
R_{NL}^k \big|_{psf} = 2 \frac{V}{n_{sat}} \sum_{k',q} C_{k' - q} X_{k+q} X_{k'} P_{k'}^* P_{k+q} P_{k'-q} + \frac{V}{n_{sat}} \sum_{k',q} C_{k'} X_{k+q} X_{k'-q} P_{k'}^* P_{k+q} P_{k'-q} .
\] (4.66)

contains additional terms providing an interaction strength due to phase space filling larger of about a factor 3 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 \(\hat{B}\hat{B}\) up to the first two terms. Schematically \(\hat{B} \to \hat{B}_B + \hat{B}_B^\dagger \hat{B}_B \hat{B}_B + \ldots\). 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 noticing that more rigourously calculated nonlinear coupling coefficients will describe more accurately parametric dynamics, as evidenced by the good quantitative agreement with experimental data, our numerical results show in [25], 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.
Chapter 5

Incoherent dynamics

The theory presented in the previous section is a flexible tool able to well describe nonlinear coherent phenomena such as pump and probe, four-wave-mixing (FWM) and parametric amplification. It represents a finer description of parametric processes occurring in coherent optical experiments with respect to the previously-used picture of weakly-interacting bosons [16, 21, 22, 78, 79]. The above coherent description is not able to take into account spontaneous or self-stimulated decay processes induced by vacuum fluctuations. In order to properly describe these phenomena light quantization has to be included [47]. Moreover, polaritons are real electronic excitations propagating in a complex interacting environment and hence subjected to scattering events determining a loss of coherence mainly originating from the interaction with lattice vibrations. For a more realistic description of the physics taking place there, we need to build up a microscopic model taking into account on an equal footing nonlinear interactions, light quantization, cavity losses and polariton-phonon interaction. To be more specific as a dominant process for excitonic decoherence in resonant emission from QWs we shall consider acoustic-phonon scattering via deformation potential interaction, whereas we shall model the losses through the cavity mirrors within the quasi-mode approach. It is worth pointing out that the approach
we are proposing may be easily enriched by several other scattering mechanisms suitable for a refinement of the numerical results.

We model QW microcavity excitons and photons in the usual way as in Ref. [20] and we are left with the semiconductor model Hamiltonian describing electronic excitations, the free cavity light modes in an ideal microcavity, plus the coupling term responsible for the polaritonic splitting. Together they provide the starting point for the description of quantum optical effects and nonlinear optics in semiconductor microcavities. In the view of the change of bases previously-mentioned, so imperative for a proper Markov calculation, we decide to treat the coupled system, described by the three Hamiltonian terms above, as our system of interest weakly interacting with the environment. Formally this means to start from the Heisenberg equations of motion resulting in Eq. (4.48) discarding the phenomenological dampings and the input term. Once obtained the polariton modes via a unitary diagonalizing transformation (we obtain real Hopfield coefficients whereas in Sec. 4.2 they are complex quantities), we apply, to the coupling of this system with the environment, the usual many-body perturbative description. We end up with the customary Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy which to the first order gives us the coherent input field, whereas to the second order the phonon and radiative scattering terms. As widely used in the literature, we shall limit ourselves up to this point thus performing a second-order Born-Markov description of the environment induced effects to the system dynamics. The details of this approach will be the subject of the next section.

Within the quasi-mode approach, the emitted light is proportional to the intracavity photon number \((t_c)\) is the transmission coefficient):

\[
I_{PL}^k(t) = t_c^2 \langle \hat{a}_k^\dagger \hat{a}_k \rangle(t) = t_c^2 \sum_i |C_{ik}|^2 \langle \hat{P}_i^\dagger \hat{P}_i \rangle(t) .
\]

(5.1)

By applying the whole machinery, when including only the lowest order terms in the input light field, the following equation for the polariton-occupations dynamics
is obtained:
\[
\frac{d}{dt} \langle \hat{P}_{ik}^\dagger \hat{P}_{ik} \rangle = -\left( \Gamma_{ph}^{i,k} + \gamma_{i,k}^{(c)} \right) \langle \hat{P}_{ik}^\dagger \hat{P}_{ik} \rangle + g_{i,k}^{(c)} + \sum_{l,k'} W_{(ik),(lk')} \langle \hat{P}_{lk'}^\dagger \hat{P}_{lk'} \rangle ,
\]
with the generation rate given by
\[
g_{i,k} = \left[ t_c C_{kp} E_{k}^{\text{in}(+)} \delta_{k,k_p} \langle \hat{P}_{i,k}^\dagger \rangle_{\text{coh}} + t_c C_{kp}^* E_{k}^{\text{in}(-)} \delta_{k,k_p} \langle \hat{P}_{i,k} \rangle_{\text{coh}} \right].
\]

The phonon-emission (+) and phonon-absorbtion (−) scattering rates read
\[
W_{(jk'),(ik)}^{\pm} = \frac{1}{\rho u S} \left| \frac{|k'-k|^2 + (q_0^z)^2}{\hbar u q_0^z} \Xi |^2 \right| \times |X_{jk'}|^2 |X_{ik}|^2 [n_{(ph)}(E_{k'}^{(ph)}(q,0^z)) + \frac{1}{2} \pm \frac{1}{2}],
\]
the 3D phonon wave vector is here expressed as \((q,0^z)\), whereas \(q_0^z\) is calculated so that the energy conservation delta function \(\delta(\hbar \omega_{jk'} - \hbar \omega_{ik} \pm E_{q,0^z}^{(ph)})\) is satisfied,
\[
\Xi = \left( D_c I_{c,h}^{\parallel}(q_z) I_{c,h}^{\parallel}(k' - k) - D_c I_{h}^{\perp}(q_z) I_{h}^{\perp}(k' - k) \right),
\]
with the overlap integrals
\[
I_{c,h}^{\parallel}(q_z) = \left[ 1 + \left( \frac{m_h}{2(m_e + m_h)} |q| a_x \right)^2 \right]^{-3/2},
I_{c,h}^{\perp}(q_z) = \int_L dz |\chi_{c,h}(z)|^2 e^{i q_z z}.
\]
We shall treat the cavity field in the quasi-mode approximation, that is to say we shall quantize the field as the mirror were perfect and subsequently we shall couple the cavity with a statistical reservoir of a continuum of external modes. This way on an equal footing we shall provide the input coherent driving mechanism (at first order in the interaction) and the radiative damping channel (within a second order Born-Markov description).

The escape rate through the two mirrors \((l \equiv \text{left}, r \equiv \text{right})\) is
\[
\gamma_{i,k}^{(c)} = \frac{2\pi}{\hbar} |C_{i,k}|^2 \sum_{s,l,T} d\omega \delta\left( \Omega_{k}^{m}[\omega] - \omega_k \right) \hbar |g_{sk}(\omega)|^2 ,
\]
and the corresponding noise term reads
\[ \Gamma^{(c)}_{i,k} = \frac{2\pi}{\hbar} |C_{i,k}|^2 \sum_{s=l,t} \int d\omega n_{sk}^m(\omega) \delta(\Omega_{sk}^m[\omega] - \omega_k) \hbar |g_{sk}(\omega)|^2, \] (5.8)
where \( \Omega_{sk}^m[\omega] \) describes the continuous spectrum of the external light modes and \( n_{sk}^m(\omega) \) is their thermal occupation, usually negligible at optical frequencies.

Within the DCTS it can be shown that \( \langle \hat{B}_{k}^{\dagger} \hat{B}_{k} \rangle \) can be developed up to the 4th order in the incident field into the sum of an exciton projector and a projector over the biexciton subspace times the proper matrix coefficient [24]. Calculating the terms from the exciton-phonon interaction under the assumption that \( \langle \hat{B}_{k}^{\dagger} \hat{B}_{k} \rangle \) is mainly due to a pure exciton population, Eq. (5.2) can be extended up to the next order in the exciting field by adding on the r.h.s. the following term:
\[ \frac{\partial}{\partial t} \langle \hat{P}_{i,k}^{\dagger} \hat{P}_{i,k} \rangle \bigg|_{NL} = 2\Re(X_{i,k} \langle \hat{P}_{i,k}^{\dagger} \hat{R}_{i,k}^{(NL)} \rangle). \] (5.9)
This last term contains a fourth-order correlator, and consequently the set of Eqs. (5.2) is no more closed. The problem can be meaningfully and significantly simplified assuming nonlinear interactions coming from a classical (coherent) input light beam. In this case, when regarding the lower polariton branch only (which will be the case of interest), we are left with two anomalous correlators:
\[ 2\Re(X_{i,k} \langle \hat{P}_{i,k}^{\dagger} \hat{R}_{i,k}^{(NL)} \rangle) = g_{k} P_{k}^2 \langle \hat{P}_{i,k}^{\dagger} \hat{P}_{2k-p-k} \rangle + + g_{k}^{*} P_{k}^2 \langle \hat{R}_{k} \hat{R}_{2k-p-k} \rangle, \] (5.10)
where
\[ g_{k} = -\frac{i N_{eff}}{N_{sat} \omega_{g}} \left( \frac{V C_{k}^*}{n_{sat} + V_{sx} X_{k}^*} \right) X_{k,p}^* X_{k} X_{2k-p-k}. \] (5.11)

## 5.1 Acoustic phonon interaction

In the case of GaAs-AlAs QW structures, the electron and hole of the excitons are considered to be well confined within a QW since the bandgap discontinuity is quite
5 – Incoherent dynamics

large. On the other hand, the lattice properties of such materials are in close proximity, thus the acoustic-phonons which interact with the quasi-two-dimensional exciton can be considered to have three-dimensional character. A quasi-two-dimensional exciton state with total in-plane center of mass (CM) wavevector $\mathbf{k}$ may be represented as [49]

$$|\lambda, \mathbf{k}\rangle = \frac{v_0}{\sqrt{A}} \sum_{\mathbf{r}_e, \mathbf{r}_h} e^{i \mathbf{k} \cdot \mathbf{R}_e} \mathbf{F}_\lambda (\mathbf{r}_e - \mathbf{r}_h; z_e, z_h) a^\dagger_{c, \mathbf{r}_e} a_{v, \mathbf{r}_h} |0\rangle, \quad \text{(5.12)}$$

$v_0$ and $A$ are the volume of the unit cell and the in-plane quantization surface, whereas $a^\dagger_{c/v, \mathbf{r}} (\hat{a}_{c/v, \mathbf{r}})$ are creation (annihilation) operator of the conduction- or valence-band electron in the Wannier representation. $\mathbf{r}_{e/h} = (\mathbf{r}_{e/h}^||, z_{e/h})$ are to be considered coordinates of the direct lattice, $|0\rangle$ is the crystal ground state and $\mathbf{R}$ the exciton center of mass coordinate $\mathbf{R} = (m_e \mathbf{r}_e^|| + m_h \mathbf{r}_h^||)/(m_e + m_h)$ with $m_e$ and $m_h$ the effective electron and hole masses. The Wannier envelope function $F_\lambda$ is normalized so that the integral over the whole quantization volume ($V = S \cdot L$) of its square modulus is equal to 1. The three-dimensional (DF) electron-phonon interaction Hamiltonian can be written as

$$H^{DF}_{e-\text{ph}} = \sum_{\mathbf{k}, \mathbf{q}} \left( \frac{\hbar |\mathbf{q}|}{2 \rho \mu V} \right)^{1/2} \left( D_c \hat{a}^\dagger_{c, \mathbf{k} + \mathbf{q}} \hat{a}_{c, \mathbf{k}} + D_v \hat{a}^\dagger_{v, \mathbf{k} + \mathbf{q}} \hat{a}_{v, \mathbf{k}} \right) \left( \hat{b}_{\mathbf{q}} + \hat{b}^\dagger_{-\mathbf{q}} \right). \quad \text{(5.13)}$$

Here $\hat{a}^\dagger_{c/v, \mathbf{k}}, \hat{a}_{c/v, \mathbf{k}}$ are creation and destruction operator of the conduction- valence-band electron in Bloch representation. Transforming from the Wannier to the Bloch representation, we shall project (5.13) into the excitonic bases. Moreover, since we are interested in the 1S exciton sector $\lambda = (n, \sigma)$ only

$$F_{1s} = W_{1s} (\mathbf{r}_e^|| - \mathbf{r}_h^||) \chi_e (z_e) \chi_h (z_h),$$

$$W_{1s} (\rho) = \sqrt{2/(\pi a_x^2)} \exp(-\rho/a_x). \quad \text{(5.14)}$$

It yields
\[ H_{\text{exc-ph}}^{DF} = \sum_{k,k',q_z} t_{k,k',q_z}^{k'} |1s,k\rangle \langle 1s,k' | \left( \hat{b}_{(k'-k,q_z)} + \hat{b}_{-(k'-k,q_z)}^\dagger \right), \quad (5.15) \]

Here

\[ t_{k,k',q_z}^{k'} = \left( \frac{\hbar \sqrt{|k' - k|^2 + q_z^2}}{2 \rho u V} \right)^{1/2} \left( D_c I_e^\dagger (q_z) I_e^\parallel (k' - k) - \right. \]

\[ \left. - D_v I_h^\dagger (q_z) I_h^\parallel (k' - k) \right). \quad (5.16) \]

It is convenient to use a fairly different form of the Hamiltonian (5.15)

\[
H_{\text{exc-ph}}^{DF} = \sum_q \left( \sum_k t_k^q |k + q\rangle \langle k| \right) \left( b_q + b_{-q}^\dagger \right) = \sum_q Q_q F_q + Q_q F_{-q}^\dagger. \quad (5.17)
\]

This form is a little bit different from (2.87) and it leads to a compact form similar to eqs. (2.100) and (2.101):

\[
\frac{d}{dt} \langle \hat{O} \rangle(t) = \langle \left[ \hat{O}, \hat{H}_0^S \right] \rangle_S - \frac{1}{\hbar^2} \sum_{q,\pm} \int_0^\infty du \left( n_{+q}^R + \theta(\pm) \right)
\]

\[ \times \left( e^{\pm \frac{q u}{\hbar}} \langle [\hat{O}, \hat{Q}_q] \hat{Q}_{-q}(-u) \rangle_S - e^{\pm \frac{q u}{\hbar}} \langle \hat{Q}_{-q}(-u) [\hat{O}, \hat{Q}_q] \rangle_S \right). \quad (5.18) \]

To exemplify our approach we shall calculate for the case \( \hat{O} = \hat{P}_{\alpha K}^\dagger \hat{P}_{\alpha K} \). Within the strong coupling region, the dressing carried by the nonperturbative coupling between excitons and cavity photons highly affects the scattering and for a microscopic calculation we are urged to leave the couple picture of (4.48) and move our steps into the polaritonic operator bases. Our aim is to produce a microscopic description of damping and fluctuation and to apply it in experiments with low and-or moderate excitation intensities, thus we expect the strong-coupling regime to become crucial in the scattering rates mainly through the polaritonic spectrum. In the spirit of the
DCTS we shall consider them as projectors over the polaritonic bases obtained form excitons and cavity modes states:

\[ |1s,k\rangle \langle 1s,k| = \sum_{i,j} X_{ik} X_{jk}^{*} |i^k\rangle \langle j^k|, \quad (5.19) \]

where it is understood we have on the left-hand side an excitonic projector and on the right-hand side a polaritonic projector. The calculation in this new symbols reads:

\[ \hat{Q}_{-q}(-u) = \sum_{k} \sum_{i,j} t_{k-q}^{i} X_{ik-q} X_{jk}^{*} |i^k-q\rangle \langle j^k| e^{-i(\omega_{k-q} - \omega_{jk})u} \quad (5.20) \]

\[
\frac{d}{dt} \langle \alpha \bar{K} | \alpha \bar{K} | \rangle_{\mu_{\eta},t} = -\frac{1}{\hbar^2} \sum_{q,\pm} \int_{0}^{\infty} du \left( n_{\pm q} \theta(\pm) \right) \left[ e^{\mp \frac{\hbar R u}{2}} \left( \sum_{i',j'} t_{k-q}^{i} t_{k}^{i'} X_{i \bar{K} - q} \right) \langle \alpha \bar{K} | | \sum_{i',j'} t_{k}^{i} t_{k-q}^{i'} X_{i' \bar{K} + q} \rangle \right] \langle \alpha \bar{K} | X_{\alpha \bar{K}} | X_{\alpha \bar{K}} | \rangle_{S} e^{-i(\omega_{\alpha \bar{K}} - \omega_{\alpha \bar{K} + q})u} + e^{\pm i \frac{\hbar R u}{2}} \left( \sum_{i',j'} t_{k-q}^{i} X_{i' \bar{K} - q} \right) \langle \alpha \bar{K} | \rangle_{S} e^{-i(\omega_{i' \bar{K}} - \omega_{j' \bar{K} + q})u} + \\
+ \sum_{i',j'} t_{k-q}^{i} X_{i' \bar{K} + q} \langle \alpha \bar{K} | \rangle_{S} e^{-i(\omega_{i' \bar{K}} - \omega_{j' \bar{K} + q})u} \quad (5.21) \]

We shall retain only the so-called T1 and T2 terms, i.e. only the secular terms in a rotating wave approximation. Using the identity

\[ \int_{0}^{\infty} e^{-i\alpha u} = \lim \frac{1}{\eta - 0} \frac{1}{i\alpha + \eta} = \pi \delta(\alpha) - i \pi(\frac{1}{\alpha}). \quad (5.22) \]
Then
\[
\frac{d}{dt} \langle | \alpha \bar{K} \rangle \langle \alpha \bar{K} | \rangle_{\mu_{int}} = -\frac{1}{\hbar} \sum_{q, \pm} \pi \left( n_{\pm q} + \theta(\pm) \right) \]
\[
\left[ \sum_{q'} \delta(\epsilon_{\alpha \bar{K}} - \epsilon_{\alpha \bar{K}_q} \mp \hbar \omega_{q}^R) t_{q-\alpha \bar{K}} | X_{\alpha \bar{K}} \rangle \langle X_{\alpha \bar{K}} | X_{\alpha \bar{K}_q} \rangle \langle X_{\alpha \bar{K}_q} | | \alpha \bar{K} \rangle \langle \alpha \bar{K} | \rangle_{S} -
\sum_{q'} \delta(\epsilon_{\alpha \bar{K}_q} - \epsilon_{\alpha \bar{K}} + \hbar \omega_{q}^R) t_{q-\alpha \bar{K}} \rangle \langle X_{\alpha \bar{K}_q} \rangle \langle X_{\alpha \bar{K}} \rangle \langle X_{\alpha \bar{K}} \rangle \langle X_{\alpha \bar{K}_q} \rangle \langle \alpha \bar{K} \rangle \langle \alpha \bar{K} | \rangle_{S} -
\sum_{q'} \delta(\epsilon_{\alpha \bar{K}} - \epsilon_{\alpha \bar{K}_q} + \hbar \omega_{q}^R) t_{q-\alpha \bar{K}} \rangle \langle X_{\alpha \bar{K}_q} \rangle \langle X_{\alpha \bar{K}} \rangle \langle X_{\alpha \bar{K}} \rangle \langle X_{\alpha \bar{K}_q} \rangle \langle \alpha \bar{K} \rangle \langle \alpha \bar{K} | \rangle_{S} +
\sum_{q'} \delta(\epsilon_{\alpha \bar{K}} - \epsilon_{\alpha \bar{K}_q} + \hbar \omega_{q}^R) t_{q-\alpha \bar{K}} \rangle \langle X_{\alpha \bar{K}_q} \rangle \langle X_{\alpha \bar{K}} \rangle \langle X_{\alpha \bar{K}} \rangle \langle X_{\alpha \bar{K}_q} \rangle \langle \alpha \bar{K} \rangle \langle \alpha \bar{K} | \rangle_{S} \right].
\]

(5.23)

Manipulating further, with the properties \( t_{q-\alpha \bar{K}} = ( t_q^\dagger )^*, n_{-q} = n_q^R, \omega_{-q}^R = \omega_q^R \), with \( \bar{k}' = \bar{k} + q^\parallel \)
\[
\frac{d}{dt} \langle \hat{P}_{\alpha \bar{K}}^\dagger \hat{P}_{\alpha \bar{K}} \rangle_{\mu_{int}} = -\Gamma_{\alpha \bar{K}}^{ph} \langle \hat{P}_{\alpha \bar{K}}^\dagger \hat{P}_{\alpha \bar{K}} \rangle_{\mu_{int}} + \sum_{l \bar{k}'} W_{(\alpha \bar{K}), (l \bar{k}')} \langle \hat{P}_{l \bar{k}'}^\dagger \hat{P}_{l \bar{k}'} \rangle_{\mu_{int}},
\]

(5.24)

with
\[
W_{(sk),(r \bar{k}')}^{\pm} = 2 \pi \hbar \sum_{q_s} | t_{r \bar{k}' q_s} \rangle \langle t_{r \bar{k}' q_s} | X_{sk} \rangle \langle X_{sk} | X_{r \bar{k}'} \rangle \langle X_{r \bar{k}'} |
\]
\[
\delta(\epsilon_{sk} - \epsilon_{r \bar{k}'} \mp \hbar \omega_{(r \bar{k}' - k q_s)}^R) \left( n_{(r \bar{k}' - k q_s)} + \theta(\pm) \right)
\]
\[
W_{(sk),(r \bar{k}')} = \sum_{\pm} W_{(sk),(r \bar{k}')}^{\pm}
\]
\[
\Gamma_{sk}^{ph} = \sum_{r \bar{k}'} W_{(r \bar{k}'),(sk)}
\]

(5.25)
5.2 The radiative scattering channel and the coherent pumping

In the spirit of the BBGKY hierarchy, one is lead to stop himself, in calculating further, to the lowest order (in the expansion) able to provide the dominant contributions. Usually such term is the second (2.93) because the first order results in a partial trace over the bath degrees of freedom of a single bath operator \( \langle b_q \rangle \) or \( \langle b_q^\dagger \rangle \) which are zero apart from a coherent state [46] – indeed the condition \( \langle b_q \rangle \neq 0 \) is one of the possible definition of coherent state$^\ast$. Modeling 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). It will be nothing but the non zero expectation value of the ensemble itself giving a non zero contribution on the 1st order. All the other incoherent bath modes will have their proper contribution in the 2nd order calculations.

It is worth noting that the treatment of the cavity losses as a scattering interaction is a result of the form chosen of the effective quasi-mode Hamiltonian. However, even if a model Hamiltonian, the quasi-mode description has given a lot of evidence as an accurate modeling tool and it is widely used in the literature.

Let us call \( R \) the quasi-mode reservoir Hamiltonian. The first order contribution to the dynamics is

\[
\frac{d\hat{\rho}}{dt}_{\mid_{\tau_{int}b}} = \hat{U}_0(t-t_0) \left( \hat{H}_{int}(t), \hat{\rho}(t) \right) \hat{U}_0^\dagger(t-t_0) = \left( \hat{H}_{int}(t), \hat{\rho}(t) \right).
\]  

(5.26)

We consider, once again, \( \hat{\rho}(t') = \hat{\rho}^S(t') \otimes \hat{\rho}^R \), we need to calculate
\[ \frac{d\hat{\rho}^S}{dt} \bigg|_{H_{int}} = tr_R \{ \frac{d\hat{\rho}}{dt} \bigg|_{H_{int}} \} = \langle \{ i\hbar \sum_{i=r,l,k} \int d\omega g_{i,k}\parallel(\omega) (\hat{a}_{k\parallel}^\dagger \hat{E}_{k\parallel} (\omega,t) \hat{\rho}^S(t') \otimes \hat{\rho}^R) \} \rangle_R + H.c. = \]

\[ = i\hbar \sum_{i=r,l,k} (\hat{a}_{k\parallel}^\dagger \{ \int d\omega g_{i,k}(\omega) \hat{E}_{k\parallel}^\dagger (\omega,t) \hat{\rho}^R \} \rangle_R \hat{\rho}^S(t') + H.c. = \]

\[ = i\hbar \sum_{i=r,l,k} \sum_{k\parallel} (\hat{a}_{k\parallel}^\dagger (g_{i,k}(\omega) \hat{E}_{k\parallel}^\dagger (\omega,t) \hat{\rho}^S(t') + H.c. ) = \]

where \( \langle g_{i,k}(\omega) \hat{E}_{k\parallel}^\dagger (\omega,t) \rangle_R \equiv \sum_p \langle E_p^{(+)}(\Omega_p,t) \rangle_R \) is the superposition of all the possible coherent pump feeds.

For the generic system operator \( \hat{O} \)

\[ \frac{d\hat{O}}{dt} \bigg|_{H_{int}} = tr_S \{ \hat{O} \frac{d\hat{\rho}^S}{dt} \bigg|_{H_{int}} \} = \]

\[ = i\hbar \sum_{i=r,l,k\parallel} (\langle g_{i,k}(\omega) \hat{E}_{k\parallel}^\dagger (\omega,t) \rangle_R \langle [\hat{O},a_{k\parallel}^\dagger] \rangle_S - \]

\[ - \langle g_{i,k}(\omega) \hat{E}_{k\parallel}^\dagger (\omega,t) \rangle_R \langle [\hat{O},a_{k\parallel}] \rangle_S . \]

The subsequent step in the BBGKY hierarchy may be obtained taking into account what has been left apart, we shall define \( \hat{E}_{k\parallel}^\dagger (\omega,t) = \hat{E}_{k\parallel}^\dagger (\omega,t) - \langle \hat{E}_{k\parallel}^\dagger (\omega,t) \rangle_R \) which represents the incoherent residual contribution not taken into account in the first order.

The quasi-mode interaction Hamiltonian reads

\[ \tilde{H}_{qm} = i\hbar \sum_{k\parallel} \int d\omega \left( g_{k\parallel}(\omega) \hat{a}_{k\parallel}^\dagger \hat{E}_{k\parallel}^\dagger (\omega,t) - \right. \]

\[ \left. - g_{k\parallel}^*(\omega) \hat{E}_{k\parallel}^\dagger (\omega,t) \hat{a}_{k\parallel} \right) . \]

90
It can be recast as (2.87) considering
\[ \hat{Q}_q = \hat{a}_{k\parallel}^\dagger ; \hat{Q}_q^\dagger = \hat{a}_{k\parallel} \]
\[ \hat{F}_q = i\hbar \sum_{i=l,r} \int d\omega g_{k\parallel}(\omega) \tilde{E}_{k\parallel}^{(-)}(\omega,t) \]
\[ \hat{F}_q^\dagger = -i\hbar \sum_{i=l,r} \int d\omega g_{k\parallel}^*(\omega) \tilde{E}_{k\parallel}^{(+)}(\omega,t) , \]
(5.29)

the calculation from eq. (2.100) is analogous to the acoustic phonon case and it yields the scattering rates eqs. (5.7,5.8).

An interesting situation occurs within the assumption of a flat quasi-mode spectrum, an approximation almost universally made in quantum optics [72]. It makes eq. (5.7) independent of the frequency:
\[ \gamma_{\alpha,k\parallel} = \sum_{i=l,r} \frac{2\pi}{\hbar} |C_{\alpha,k\parallel}|^2 |h|g_{i,k\parallel}|^2 = |C_{\alpha,k\parallel}|^2 \sum_{i=l,r} \gamma_{i,k\parallel}^{(m)} , \]
(5.30)
where \( \gamma_{i,k\parallel}^{(m)} \) is the (i-side) damping of the cavity without the quantum well.

Thus
\[ \sum_{i=l,r} \gamma_{i,k\parallel}^{(m)} = 2\pi \sum_{i=l,r} |g_{i,k\parallel}|^2 , \]
(5.31)
there are two situations:

- equal damping: \( \gamma_{l,k\parallel}^{(m)} = \gamma_{r,k\parallel}^{(m)} \), we can define the transmission coefficient of the i-side
\[ |g_{i,k\parallel}|^2 = \frac{\gamma_{i,k\parallel}^{(m)}}{2\pi} \equiv t_{c,i}^2 \]
(5.32)

- we know the ratio:

\[ \begin{cases} R = \frac{\gamma_{r,k\parallel}^{(m)}}{\gamma_{l,k\parallel}^{(m)}} \\ \gamma_{\text{tot},k\parallel}^{(m)} = \gamma_{r,k\parallel}^{(m)} + \gamma_{l,k\parallel}^{(m)} \end{cases} \Rightarrow \begin{cases} \gamma_{l,k\parallel}^{(m)} = \frac{1}{1+R} \gamma_{\text{tot},k\parallel}^{(m)} \\ \gamma_{r,k\parallel}^{(m)} = \frac{R}{1+R} \gamma_{\text{tot},k\parallel}^{(m)} \end{cases} \]
The transmission coefficients follow. In the light of the definition of $t_{(c),i}^2$, it becomes evident that the semiclassical coherent input feed could also be modeled from the beginning with an effective Hamiltonian:

$$H_p = i\hbar \sum_{k\parallel} E_{k\parallel} (\hat{a}_{k\parallel}^\dagger - \hat{a}_{k\parallel}), \quad (5.33)$$

where (the $C$-number) $E_{k\parallel} = \sum_p t_{(c),p} \langle E_p^{(-)}(\Omega_p,t) \rangle_R$ represents the incoming coherent input beams [20].
Chapter 6

Nonequilibrium quantum Langevin approach to parametric emission

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 operators differential equations, and above all, for its invaluable flexibility and strength in performing even multitime correlation calculations, so important when dealing with quantum correlation properties of the emitted light. Moreover, as we will 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 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 bath operators initial values which, due to the impossibility to know such values, are considered noise sources of a stochastic nature. Normally the model considered is of the form of harmonic oscillators coupled linearly to a bosonic environment. The standard
statistical viewpoint is easy 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 either. In 1966 Melvin Lax, with clear in mind the lesson of classical statistical mechanics of Brownian motion, in Ref. [64] proposed for the first time that as soon as one is left with a closed set of equations for the mean motion they can be promoted to equations for the global bare operators provided to consider additive noise sources endowed by the proper statistics due to the system dynamics. He showed that in a Markovian environment they must fulfill generalized Einstein equations which are a sort of time dependent non-equilibrium fluctuation-dissipation theorem. In order to be more specific, let us consider a single semiclassical pump feed resonantly exciting the lower polariton branch at a given wavevector \( k_p \). The nonlinear term \( R_{NL} \) couples pairs of wavevectors, let’s say \( k \), the signal, and \( k_i = 2k_p - k \), the idler. The Heisenberg-Langevin equations for the generic couple read

\[
\begin{align*}
\frac{d}{dt} \hat{P}_k &= -i\tilde{\omega}_k \hat{P}_k + g_k \hat{P}^\dagger_{k_i} \hat{P}^2_{k_p} + \hat{F}_k \\
\frac{d}{dt} \hat{P}^\dagger_{k_i} &= i\tilde{\omega}_{k_i} \hat{P}^\dagger_{k_i} + g_{k_i}^* \hat{P}_k \hat{P}^2_{k_p} + \hat{F}^\dagger_{k_i},
\end{align*}
\]

(6.1)

where \( P_{k_p} \) is the classical pump feed, the renormalized complex polariton dispersion \( \tilde{\omega}_k \) include the effects of relaxation and pump-induced renormalization, \( \tilde{\omega}_k = \omega_k - i\Gamma_k^{(tot)} / 2 + h_k |P_{k_p}|^2 \), with \( \Gamma_k^{(tot)} = (\Gamma_k^{(ph)} + \gamma_k^{(c)}) \), and

\[
h_k = \frac{1}{N_{eff}} \left( \frac{V}{n_{sat}} C^*_{k_p} X_{k_p} |X_k|^2 + \frac{V}{n_{sat}} C^*_{k} X_k |X_{k_p}|^2 \right)
+ 2V_{xx} |X_{k_p}|^2 |X_k|^2.
\]

(6.2)

The nonlinear interaction terms in Eq. (6.1) determine a pump-induced blue-shift of the polariton resonances and a pump-induced parametric emission. In Eqs. (6.1) 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 [66] that both effects depends 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.

The Langevin noise source operators are such that their expectation values $\langle \hat{F}_N \rangle$ vanish, but their second order moments do not [64]. They are intimately linked up with the global dissipation and in a Markovian environment they take the form:

$$\langle \hat{F}_M(t) \hat{F}_N(u) \rangle = 2 \langle \hat{D}_{MN} \rangle \delta(t-u), \quad (6.3)$$

where

$$2 \langle \hat{D}_{MN} \rangle = \frac{d}{dt} \langle \hat{M} \hat{N} \rangle - \langle \left\{ \frac{d}{dt} \hat{M} \right\} \hat{N} \rangle - \langle \hat{M} \left\{ \frac{d}{dt} \hat{N} \right\} \rangle, \quad (6.4)$$

$$\left\{ \frac{d}{dt} \hat{N} \right\} \equiv \frac{d}{dt} \hat{N} - \hat{F}_N, \quad (6.5)$$

where $\hat{M}, \hat{N}$ are generic operators.

Equation (6.3) is an (exact) time dependent Einstein equation representing a fluctuation-dissipation relation valid for nonequilibrium situations, it witnesses the fundamental correspondence between dissipation and noise in an open system. For the sake of clarity we are considering in the single $\langle \rangle$ the partial trace over the bath together with the other trace over the system degrees of freedom, indeed in Eq. (6.3) and Eq. (6.4) we are facing C-numbers. However the general theory has deeper consequences and the most generic form of Eq. (6.3) would involve the operators $\hat{M}$ and $\hat{N}$ as system operators traced out of the bath degrees of freedom. In this guise it assumes the most evocative form of

$$2 \hat{D}_{MN} = \frac{d}{dt} \left( \hat{M} \hat{N} \right) - \left( \left\{ \frac{d}{dt} \hat{M} \right\} \hat{N} \right) - \left( \hat{M} \left\{ \frac{d}{dt} \hat{N} \right\} \right), \quad (6.6)$$
hence, $\hat{D}_{MN}$ 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 (6.3) and Eq. (6.4) make the resulting "fluctuation-dissipation" relations between $\hat{D}_{MN}$ 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. Apart from these formal arguments, we shall be interested in applying Langevin coefficients to calculate $\langle \hat{P}_k^\dagger \hat{P}_k \rangle$ and hence we shall need

$$2\langle \hat{D}_{P_k^\dagger R_k} \rangle = \frac{d}{dt} \langle \hat{P}_k^\dagger \hat{P}_k \rangle - \langle \left\{ \frac{d}{dt} \hat{P}_k^\dagger \right\} \hat{P}_k \rangle - \langle \hat{P}_k^\dagger \left\{ \frac{d}{dt} \hat{P}_k \right\} \rangle,$$

(6.7)

and the diffusion coefficient for the two operators in reverse order. It is worth noticing that thanks to this structure all the coherent contributions cancel out automatically giving us an easy way to separate coherent and incoherent parts, but at the same time to treat them on an equal footing when calculating the final result. For instance let us explicitly calculate (6.7):

$$2\langle \hat{D}_{P_k^\dagger R_k} \rangle(t) = \sum_{k'} W_{k,k'} \langle \hat{P}_{k'}^\dagger \hat{P}_{k'} \rangle(t) + \Gamma_{kk'}^c.$$  

(6.8)

Inspecting Eq. (5.2) and Eq. (5.10), it results that in the low and intermediate excitation regime the main incoherent contribution to the dynamics is the PL the pump produces by itself, the effects on the PL of subsequent pump-induced repopulation arising from the nonlinear parametric part is negligible, that is to say the occupancies of the couple signal-idler are at least one order of magnitude smaller than the pump occupancy. This means that in (6.8) we can consider at the right
hand side the solution in time of Eq. (5.2), i.e. only incoherent lowest order contributions. The other important diffusion coefficient reads:

$$
2\langle \hat{D}_{\hat{P}_k}^\dagger \hat{P}_k \rangle (t) = \sum_{k'} W_{k,k'} \langle \hat{P}_{k'}^\dagger \hat{P}_k \rangle (t) + \Gamma_k + \gamma_k = \sum_{k'} W_{k,k'} ( \langle \hat{P}_{k'}^\dagger \hat{P}_k \rangle (t) + 1 ) + \Gamma_k + \gamma_k .
$$

(6.9)

In the following we shall indicate as \( \langle \hat{P}_k^\dagger \hat{P}_k \rangle_{PL} \) this solution representing the polariton occupation of the pump-induced PL.

The general solution of Eqs. (6.1) in the pump reference frame reads:

$$
P(t) = e^{\int_0^t M(t') dt'} P(0) + \int_0^t e^{\int_0^t M(t'') dt''} \mathcal{K}(t') dt'
$$

(6.10)

$$
P(t) = \begin{pmatrix} \hat{P}_k(t) \\ \hat{P}_{2k_p-k}^\dagger(t) \end{pmatrix}, \mathcal{K}(t) = \begin{pmatrix} \hat{F}_{P_k}(t) \\ \hat{F}_{P_{2k_p-k}}^\dagger(t) \end{pmatrix}
$$

$$
M = \begin{pmatrix} \bar{\omega}_k & \Delta(k,\tau) \\ \Delta^*(k,\tau) & \bar{\omega}_{2k_p-k} \end{pmatrix},
$$

where

$$
\bar{\omega}_k = -\omega_k ,
$$

$$
\bar{\omega}_{2k_p-k} = -i(\omega_{2k_p-k} - 2\omega_{k_p}) ,
$$

the pump is \( \hat{P}_{k_p} = \hat{P}_{k_p}^0 e^{-i\omega_{k_p}^0 t} \),

$$
\hat{P}_{2k_p-k}^\dagger = \hat{P}_{2k_p-k}^\dagger e^{-i2\omega_{k_p}^0 t} ,
$$

$$
\hat{F}_{P_{2k_p-k}}^\dagger = \hat{F}_{P_{2k_p-k}}^\dagger e^{-i2\omega_{k_p}^0 t} ,
$$

$$
\omega_{k_p}^0 = \omega_k + h_k |\mathcal{P}_{k_p}|^2 ,
$$

$$
\Delta(k,\tau) = g_k \mathcal{P}_{k_p}^0 e^{-i2\omega_{k_p}^0 \tau} ,
$$

(6.11)

Equation (6.10) can be written in a more explicit form by exploiting the following
identity:
\[
e^{\int_{t_1}^{t_2} A(t) dt} = \alpha_1(t_1, t_2) \int_{t_1}^{t_2} A(t) dt + \alpha_0(t_1, t_2) \mathbb{I}
\]
\[
\alpha_0(t_1, t_2) = \frac{\Lambda_+(t_1, t_2) e^{\Lambda_-(t_1, t_2)} - \Lambda_-(t_1, t_2) e^{\Lambda_+(t_1, t_2)}}{\Lambda_+(t_1, t_2) - \Lambda_-(t_1, t_2)}
\]
\[
\alpha_1(t_1, t_2) = \frac{e^{\Lambda_+(t_1, t_2)} - e^{\Lambda_-(t_1, t_2)}}{\Lambda_+(t_1, t_2) - \Lambda_-(t_1, t_2)}
\]
\[
\Lambda_{\pm}(t_1, t_2) = \int_{t_1}^{t_2} \lambda_{\pm}(\tau) d\tau
\]
\[
\int_{t_1}^{t_2} \text{diag}[A(t)] dt = \begin{pmatrix} \Lambda_-(t_1, t_2) & 0 \\ 0 & \Lambda_+(t_1, t_2) \end{pmatrix}
\]
\[
\lambda_{\pm} = w^* \pm \sqrt{w^* + \Delta^2}
\]
\[
w^* = \frac{\omega_k^* + \omega_{2k-p-k}^*}{2}, \quad w^- = \frac{\omega_k - \omega_{2k-p-k}^*}{2}.
\]

(6.12)

**polariton occupation dynamics**

Eq. (6.10) with Eq. (6.12) provides an easy and general starting point for the calculation of multi-time correlation functions which are key-quantities in quantum optics. Taking the expectation values of the appropriate products it yields

\[
\langle \hat{P}_k \hat{P}_k \rangle = |c_1(0, t)|^2 N_k(0) + |c_2(0, t)|^2 (N_{2k-p-k}(0) + 1) + \int_0^t d\tau |c_1(\tau, t)|^2 2 \langle \hat{D}_{k} \hat{P}_k \rangle (\tau) + \int_0^t d\tau |c_2(\tau, t)|^2 2 \langle \hat{D}_{p_k-k} \hat{P}_k \hat{P}_{2k-p-k} \rangle,
\]

here

\[
c_1(t_1, t_2) = \alpha_1(t_1, t_2) \int_{t_1}^{t_2} d\tau (\frac{\Gamma_k^{(tot)}}{2} - i\omega_k^{(h)}) + \alpha_0(t_1, t_2)
\]
\[
c_2(t_1, t_2) = \alpha_1(t_1, t_2) \int_{t_1}^{t_2} d\tau \Delta(k, \tau).
\]

(6.13)

In all the situations under investigation, the thermal population of photons at optical frequencies are negligible, hence \(\Gamma_k^{(c)} \approx 0\). In Eq. (6.13) it is evident the great flexibility of the Langevin method, even in single-time correlations. It represents
a clear way to "decouple" the incoherent and the coherent dynamics in an easy
and controllable fashion. In the important case of steady-state, where the standard
Langevin theory could at least in principle be applied, we have nonequilibrium
Langevin sources which become:

\[
0 = -\Gamma_k^{(\text{tot})} \langle \hat{P}_k^\dagger \hat{P}_k \rangle + \sum_{k'} W_{k,k'} \langle \hat{P}_{k'}^\dagger \hat{P}_k \rangle + \Gamma_c^c \\
2 \langle \hat{D}_{k'} \hat{P}_k \rangle = \sum_{k'} W_{k,k'} \langle \hat{P}_{k'}^\dagger \hat{P}_k \rangle (t) + \Gamma_k^c \\
0 = -\Gamma_k^{(\text{tot})} \langle \hat{P}_k \hat{P}_k^\dagger \rangle + \sum_{k'} W_{k,k'} \langle \hat{P}_k \hat{P}_{k'}^\dagger \rangle + \Gamma_k^c + \gamma_k^c \\
2 \langle \hat{D}_{k'} \hat{P}_k \rangle = \sum_{k'} W_{k,k'} \langle \hat{P}_k \hat{P}_{k'}^\dagger \rangle + \Gamma_k^c + \gamma_k^c ,
\]
giving

\[
2 \langle \hat{D}_{k'} \hat{P}_k \rangle (t) = \Gamma_k^{(\text{tot})} \langle \hat{P}_{k'}^\dagger \hat{P}_k \rangle (t) \\
2 \langle \hat{D}_{k'} \hat{P}_k \rangle (t) = \Gamma_k^{(\text{tot})} (\langle \hat{P}_{k'}^\dagger \hat{P}_k \rangle (t) + 1) ,
\]
i.e. the standard statistical viewpoint is recovered in steady-state. Moreover, in the
limit of pump intensity tending to zero it is the PL which governs the dynamics.
Indeed Eq. (5.2) in this situation reads

\[
\frac{d}{dt} \langle \hat{P}_k^\dagger \hat{P}_k \rangle = -\Gamma_k^{(\text{tot})} + \langle \hat{P}_k^\dagger \hat{P}_k \rangle + \sum_{k'} W_{k,k'} \langle \hat{P}_{k'}^\dagger \hat{P}_k \rangle ,
\]
when, at least formally, we consider the right-hand side as known we can integrate,
obtaining

\[
\langle \hat{P}_k^\dagger \hat{P}_k \rangle = \int_0^t dt' e^{-\Gamma_k^{(\text{tot})}(t-t')} \sum_{k'} W_{k,k'} \langle \hat{P}_{k'}^\dagger \hat{P}_k \rangle
\]
which is the limit of excitation intensity to zero of Eq. (6.13). The form of \(2 \langle \hat{D}_{k'} \hat{P}_k^\dagger \rangle\) guarantees this fact for the reverse order calculation.

Furthermore, standard Langevin theory gives some problems in dealing with in-
teraction forms more complicated than the standard linear two-body coupling and
some additional approximations are needed. Lax technique, on the contrary, provides us with the correct Langevin noise sources in the generic nonequilibrium case no matter of the operatorial form of the reservoir (weak) interaction Hamiltonian to implement. They properly recover the well-known steady-state result even if they depend, in the generic case, on the scattering rates rather than on dampings, on the contrary to standard Langevin description.

time-integrated spectrum

The spectrum of a general light field has always been of great interest in understanding the physical properties of light. Any spectral measurement is made by inserting a frequency-sensitive device, usually a tunable linear filter, in front of the detector. What is generally called “spectrum” of light is just an appropriately normalized record of the detected signal as a function of the frequency setting of the filter [73]. Here we are interested to the power spectrum of a quantum-field originating from pulsed excitation and thus not at steady state. The time-integrated spectrum of light for a quantum-field can be expressed as [74, 73].

$$I_k(\omega, T) = \kappa \frac{2\Gamma}{T - t_0} \int_{t_0}^{T} dt_1 \int_{t_0}^{T} dt_2 \langle \hat{E}^{(-)}_{k}(t_1) \hat{E}^{(+)}_{k}(t_2) \rangle e^{-(\Gamma + i\omega)(T-t_1)} e^{-(\Gamma - i\omega)(T-t_2)} ,$$

(6.14)

where $\Gamma$ is the bandwidth of the spectrometer (e.g. of the Fabry-Perot interferometer) and $\hat{E}^{(-)}_{k}$ ($\hat{E}^{(+)}_{k}$) are the field operators corresponding to the light impinging on the detector, $\kappa$ is nothing but a proportional factor depending on the detector parameters and efficiency. Within the quasi-mode approach [72, 75] the spectrum of transmitted light is proportional to the spectrum of the intracavity field. In our situation, in the very narrow bandwidth limit and considering a beam with given in-plane wave vector, [76] it reads

$$I_k(\omega, T) = \kappa \frac{t_e^2}{T - t_0} \int_{t_0}^{T} dt_1 \int_{t_0}^{T} dt_2 \langle \hat{a}^{\dagger}_{k}(t_1) \hat{a}_{k}(t_2) \rangle e^{-i\omega(t_1-t_2)} .$$

(6.15)
By expressing the cavity-photon operator in terms of polariton operators, one obtains
\[ I_k(\omega, T) = \frac{\kappa t_c^2}{T - t_0} \int_{t_0}^{T} dt_1 \int_{t_0}^{T} dt_2 \sum_i |C_{ik}|^2 \langle \hat{P}_{ik}^\dagger(t_1) \hat{P}_{ik}(t_2) \rangle e^{-i\omega(t_1 - t_2)}. \] (6.16)

In our experimental conditions the upper polariton contribution is negligible, thus we need to calculate
\[ I_k(\omega, T) = \frac{\kappa t_c^2 |C_k|^2}{T - t_0} \int_{t_0}^{T} dt_1 \int_{t_0}^{T} dt_2 \langle \hat{P}_{k}^\dagger(t_1) \hat{P}_{k}(t_2) \rangle e^{-i\omega(t_1 - t_2)}. \] (6.17)

By using Eq. (6.10) and the properties of noise operators (6.3), one obtains:
\[
\langle \hat{P}_{k}^\dagger(t_1) \hat{P}_{k}(t_2) \rangle = c_1(0,t_1)^*c_1(0,t_2)N_k(0) + c_2(0,t_1)^*c_2(0,t_2)(N_{2k_p-k}(0) + 1) + \\
+ \delta_{t_{\alpha, \min(t_1,t_2)}} \int_0^{t_{\alpha}} d\tau \ c_1(\tau,t_1)^*c_1(\tau,t_2) \sum_{k'} W_{k,k'} \langle \hat{P}_{k'}^\dagger \hat{P}_{k'} \rangle_{\text{PL}}(\tau) + \\
+ \delta_{t_{\alpha, \min(t_1,t_2)}} \int_0^{t_{\alpha}} d\tau \ c_2(\tau,t_1)^*c_2(\tau,t_2) \left( \sum_{k'} W_{2k_p,k-k'} \left( \langle \hat{P}_{k'}^\dagger \hat{P}_{k'} \rangle_{\text{PL}}(\tau) + 1 \right) + \gamma_{2k_p-k}^{(c)} \right) .
\] (6.18)

### 6.1 Non-classical properties of microcavity polaritons: analytical study

In order to obtain some information about the nonclassical features of the emitted light (and hence of the internal dynamics) we consider coincidence detections in Ref. [77]. In particular entangled photon pairs display forth order interference in the joint detection probability that cannot be obtained in classical systems. Interference visibility is defined as
\[ V_k = \frac{2\langle \hat{N}_{sk} \hat{N}_{2k_p-k} \rangle}{\langle \hat{N}_{sk}^2 \rangle + \langle \hat{N}_{2k_p-k}^2 \rangle + 2\langle \hat{N}_{sk} \hat{N}_{2k_p-k} \rangle}, \] (6.19)

where \( \langle \hat{N}_{sk} \rangle = \langle \hat{P}_{sk}^\dagger \hat{P}_{sk} \rangle \) (\( :: \) stands for normal order and \( k_i = 2k_p - k \)). In the presence of classical beams we will see that a small visibility can be produced by
classical correlations, however it cannot exceed $1/2$, this way a visibility detection measurement above this value has to be considered a clear signature of a quantum behaviour.

So far things has been done analytically, but as soon as we obtained the various equations we figured out that microscopic rigorous calculations need hard numerical simulations of the full dynamics of the quantum system. For the present purpose we shall exploit once again the Heisenberg-Langevin approach that allows us to grant the correctness of the equations without, strictly speaking, to join them with the microscopic calculations of the damping terms. Indeed, once the equations for the mean motions are set, whether or not the parameters are microscopic or phenomenological or taken from the experiment does not matter, all the theoretical apparatus of noise sources is still valid. For the sake of simplicity we shall consider here the balanced case ($\Gamma_{s/i}^{(\text{tot})} = \Gamma$) with continuous single-pump excitation, we then obtain analytical simple expressions (the pump is $P_{kp} = P^o_{kp} e^{-i \omega_{kp} t}$ and the idler $\tilde{P}_{i,2kp-k}^\dagger = \tilde{P}_{i,2kp-k}^\dagger e^{-i 2\omega_{kp} t}$):

\[
\begin{pmatrix}
\hat{P}_{sk}(t) \\
\tilde{P}_{i,2kp-k}(t)
\end{pmatrix} = e^{-\int_0^t M(t') dt'} \begin{pmatrix}
\hat{P}_{sk}(0) \\
\tilde{P}_{i,2kp-k}(0)
\end{pmatrix} + \int_0^t e^{-\int_{t'}^t M(t'') dt''} K(t') dt'
\]

\[M = \begin{pmatrix}
\Gamma_{s}^{(\text{tot})} + i \omega_{k}^P & -\kappa P^o_{kp} \\
-\kappa^* P_{kp}^{o2} & \Gamma_{i}^{(\text{tot})} + i(\omega_{2kp-k}^P - 2\omega_{kp}^P)
\end{pmatrix}
\]

\[K = \begin{pmatrix}
\hat{F}_{psk} \\
\tilde{F}_{i,2kp-k}^\dagger
\end{pmatrix}.
\]

We shall solve diagonalizing $M$, and developing $\hat{P}_{sk}(t)$ and $\tilde{P}_{i,2kp-k}(t)$ in terms of the corresponding eigenvector. In steady-state with phase-matching it yields:

\[V_k = \frac{4N^2_{P}^{(2\text{th})} (1 + \chi)}{4N^2_{P}^{(2\text{th})} (3 + \chi) + 16N^2_{P}^{(2\text{th})} \chi^2 + (1 + 3\chi^2) \chi^2},
\]

being $\chi = \Delta/\Gamma$, $\Delta = |\kappa P^o_{kp}|$, the effective interaction parameter and $N^2_{P}^{(2\text{th})}$ is the noise due to photoluminescence. According to Fig. 6.1 in the absence of noise
and at low excitation intensity $\Delta \to 0$ and $V \to 1$: a clear fingerprint of the quantum property of the emitted light; the other limiting situation is near the threshold where $\chi \to 1$, thus $V \to 1/2$, the classical value. In the limit of low excitation, as soon as some small amount of noise is present, it spoils the system of its correlation, but even at moderate excitation intensity the non-linear processes act properly introducing a relative resistance to noise thus showing a parameter region where feasible experiments may be performed.

Figure 6.1. (color online). Contour plot of the steady-state visibility $V_k$ for common values of polariton noise and normalized coupling $\chi$. 
6.2 Photoluminescence dynamics: numerical results

In numerical calculations we need to discretize in $k$-space. Although, thanks to confinement, cavity photons acquire a mass, this is about 4 order of magnitude smaller than the typical exciton mass, thus the polariton splitting results in a very steep energy dependence on the in-plane wavevector near $k = 0$ ($k = \omega \sin \theta / c$) (see Fig. 6.2). This very strong variation of the polariton effective mass with momentum makes difficult the numerical integration of the polarion PL rate-equations (5.2). For example if PL originates from a pump beam set at the magic-angle (see Fig. 6.2) or beyond, a small temperature of 5 K is sufficient to enable scattering processes towards states at quite higher $k$-vectors, thus it is necessary to include a computational window in $k$-space, significantly beyond $k_{\text{pump}}$. Usually, in finite volume numerical calculations, the $k$-space mesh is chosen uniform, but a dense grid suitable for the strong coupling region would result in a grid of prohibitively large number of points for (e.g. thermally activated) higher $k$, on the other hand a mesh well-suited for polaritons at higher $k$-values would consist of so few points close to $k = 0$ to spoil the results gathered from the numerical code completely of their physical significance. Following Ref. [78] we choose a uniformly spaced grid in energy which results in an adaptive $k$-grid (in modulus), in addition, thanks to the rotating symmetry of the dispersion curve, we choose a uniformly distributed mesh in the angle $\theta$ so that $k = (k, \theta)$.

Following Ref. [78] we chose a uniformly spaced grid in energy, defined as

$$E_i = E(k = 0) + i \Delta E,$$

with $i = 0, 1, 2, \ldots$. $\Delta E$ defines the energy spacing, as Ref. [78] a good value could be $\Delta E = 5 \cdot 10^{-2}$meV, we chose in our simulation to freely set the maximum value of the
wavevector and of the number of point in the mesh, \( \Delta E \) follows consistently (anyway we took care to have value less or equal to \( 5 \cdot 10^{-2} \text{meV} \)). This uniformly distributed energy grid results in an adaptive \( k \)-grid (in modulus), in addition, thanks to the rotating symmetry of the dispersion curve, we chose a uniformly distribute mesh in the angle \( \theta \) so that \( k = (k, \theta) \). Our aim was to model situation with more than one coherent pump feed, thus we decided to consider explicitly the \( \theta \) dependence. Anyway, for the typical times considered the net result we find is a rotational invariant photoluminescence pattern. Rather surprisingly, even if this adaptive choice allows for a numerical integration of our rate equation (5.2), it carries unbearable approximations for the parametric process (6.13). This is due to the different features the parametric phenomenon has with respect to pump photoluminescence. The latter arises form the balance of competing incoherent scattering events connecting different regions in \( k \)-space, on the contrary the numerical grid enters parametric simulations only in the choice of any signal-idler couple. It is worth underlining that our method enables, under certain assumptions, the (computationally advantageous) decoupling of incoherent dynamics from parametric processes allowing us to make the proper choices for the to competing contributions whenever needed.

In particular, we seed the system at a specific \( k \) and first of all we calculate the pump-induced PL by means of eq. (5.2). Because of the very steep dispersion curve and the large portion of \( k \)-space to be taken into account, in the numerical solution we need to exploit the adaptive grid above mentioned. Afterwards we use this pump-induced PL, \( \langle \hat{P}_k^\dagger \hat{P}_k \rangle_{\text{PL}} \), as a known input source in eq. (6.13) where it is largely more useful to discretize uniformly in \( k \).

We consider a SMC analogous to that of Refs. [17, 14] consisting of a 25 nm GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As single quantum well placed in the center of a \( \lambda \) cavity with AlAs/Al\(_{0.15}\)Ga\(_{0.85}\)As Bragg reflectors. The lower polariton dispersion curve is shown in Fig. 1. The simulations are performed at \( T = 5 \text{K} \) and the measured cavity
Figure 6.2. (color online). Energy dispersion of the lowest polariton branch for the structure of Ref. [14] consisting of a 25 nm GaAs/Alₐ₄Gaₐ₇As single quantum well placed in the center of a λ cavity with AlAs/Al₀.₁₅Ga₀.₈₅As 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 \( k = 0 \) and \( k = 2k_m \). The red curve symbolizes incoherent pump scattering at \( T = 0 \) K, e.g. due to acoustic phonon interaction. Because of the very steep dispersion curve in the strong coupling region due to strong coupling, for a pump beam set at the magic angle or beyond, even a small temperature of 5 K is sufficient to enable scattering processes towards states at quite higher \( k \)-vectors, thus it becomes necessary to include a computational window in k-space, significantly beyond \( k_{pump} \) making difficult numerical simulations.

Linewidth is \( h\gamma_c = 0.26 \) meV. The laser pump is modeled as a single Gaussian-shaped impulse of FWHM \( \tau = 1 \) ps exciting a definite wave vector \( k_p \) 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 \( \mu \)m\(^-2\) per pulse. We observe that Ref. [14] excites with a linearly polarized laser whose intensities \( I^L \) are multiple of an \( I^L_0 \) corresponding to a photon flux of 21 \( \mu \)m\(^-2\) per pulse too. In situations where the PSF and the MF terms dominate the nonlinear parametric interaction, there is no polarization mixing.
and two independent parametric processes take place, the first involving circularly polarized modes only and the second involving counter-circularly polarized modes. So for comparison with theory the effective density in those experiments is half the exciting density: \( I = I_L/2 \).

It has been theoretically shown [79], that it is quite difficult to populate the polaritons in the strong coupling region 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 [80]. This is mainly due to other more effective scattering mechanisms [81] 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 [81]. 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 in the mean agree with experimental values.

We now present the results of numerical solutions of Eq. (6.13) taking into account self-stimulation but neglecting the less relevant pump-induced renormalization
of polariton energies [25].

![Figure 6.3](image.png)

Figure 6.3. Calculated time dependent polariton mode-occupation at \( k = (0,0) \) obtained at four different pump intensities in comparison with the time dependent pump-induced PL at the corresponding \( k \). The laser pump is modeled as a single Gaussian-shaped impulse of FWHM \( \tau = 1 \) ps exciting a definite wave vector \( k_p = (k_m,0) \) and centered at \( t = 4 \) ps. The calculated values are in good quantitative agreement with the measured value of Ref. [14], with no fitting parameter needed nor exploited.

Figures 6.3 and 6.4 show the calculated time dependent polariton mode-occupation of a signal-idler pair at \( k = (0,0) \) and at \( k = (2k_m,0) \), respectively, obtained for four different pump intensities in comparison with the time dependent pump-induced PL at the corresponding \( k \). The pump beam is sent at the magic angle [12] \( (k_m \simeq 1.44 \cdot 10^6 \text{m}^{-1}) \) 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. 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. 6.3 and 6.4 we show the calculated polariton occupation, i.e. the number of polaritons per mode.

![Figure 6.4. Calculated time dependent polariton mode-occupation at $k = (2k_m, 0)$. It has been obtained under the same condition as Fig. 6.3. It is clear the different pump-induced PL dynamics with respect to Fig. 2, specifically a residual queue at high time values.](image)

In our calculations no fitting parameter is needed, nor exploited (apart from the doubling of the phonon scattering rates). Moreover our results predict in good agreement with the experimental results of Ref. [14] the pump intensity at which parametric scattering, superseding the pump-PL, becomes visible. It is clear the different pump-induced PL dynamics of the mode-occupation at $k_x = 2k_m$ (Fig. 6.4) with respect to that at the bottom of the dispersion curve of Fig. 6.3. Specifically a residual queue at high time values, due to the very low radiative decay of polaritons with $k$-vectors beyond the inflection point, can be observed. Furthermore we notice
that already at moderate pump excitation intensities the parametric contribution dominates. It represents a clear evidence that we may device future practical experiments exploiting such a window where the detrimental pump-induced PL contribution is very low meanwhile we face a good amount of polaritons per mode. Indeed for photon-counting coincidence detections to become a good experimental mean 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. Our results clearly show that there is a practical experimental window where we would address a situation where all these conditions would be well fulfilled.

We now focus our attention on the positive part of the $k_y = 0$ section at different pump powers. In Fig. 6.5 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 $I = 10 I_0$ ($I^L = 20 I_0$). 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. [12] and Ref. [82]. However, at so high $k_x$ values the photon component is very small and even if the polariton idler occupation is very high (as the inset if Fig. 6.5 shows), the outgoing idler light is so weak to give some difficulties in real experiments [14]. Moreover we can notice that the parametric process removes the phonon bottleneck in the region close to $k = 0$. An analogous situation occurs also in Ref. [80], though with a different SMC, where it can be seen the bottleneck removal in $k = 0$ due to the parametric emission.

Fig. 6.6 shows the impact on the time integrated patterns of the calculated pump-induced PL. We consider, for different excitation intensities, the solutions of Eq. (6.13) with and without the pump-induced PL occupations. As can be seen its
Figure 6.5. Time-integrated outgoing photon emission intensity. The pump is set at $k_p = (k_m, 0)$. 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 $I^L = 20 I_0$. Moreover the parametric process removes the phonon bottleneck in the region closed to $k = 0$. As expected the specific signal-idler parametric scattering with the signal in $k_x = 0$ and the idler in $k_x = 2k_m$ is favourite and at higher pump intensities dominates the light emission. 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.

inclusion does not result in an uniform noise background, but it seems to somewhat remember its incoherent nonuniform distribution (the one depicted in the corresponding curve, i.e. PL, in Fig. 6.5). As can be clearly gathered from the figure, the pump-induced PL has a non negligible contribution in a region in $k$-space resonant for the parametric processes. As a consequence at intermediate excitation intensities it adds up to the parametric part reaching a contribution even comparable to the peak of emission set in $k = 0^+$. Only beyond the above mentioned
The impact on the time integrated patterns of the calculated pump-induced PL for different excitation intensities is shown (the pump is set in \( k_p = (k_m,0) \)). Here the solutions of Eq. (6.13) with and without the pump-induced PL occupations are depicted. On the contrary to what implicitly considered in previous phenomenological theories it results in a non-uniform noise background and hence its momentum distribution has to be included for a realistic microscopic calculation of the emission patterns. Moreover it is non-negligible in a region in \( k \)-space resonant for the parametric processes and hence at intermediate excitation intensities it adds up to the parametric part reaching a contribution even comparable to the peak of emission set in \( k = 0^+ \) up to the threshold around \( I^t = 20 I_0 \). In the inset the ratio of the homogenous solution with the global emission at \( k = 0^+ \), calculated without \( \langle \hat{P}_k \hat{A}_k \rangle_{PL} \), is depicted. For increasing pump intensities the two contributions (homogeneous and particular) in Eq. (6.13) still display comparable contributions, hence for a proper description of the spontaneous parametric emission they must be both included.

threshold the parametric emission is able to take over pump-induced PL and results in the great emission in the bottom of the dispersion curve of Ref. [12].

These results clearly show that PL emission does not become negligible at quite high excitation densities, but, being amplified by the parametric process, determines a redistribution of polariton emission displaying qualitative differences respect to calculations neglecting PL. An interesting question regarding these phenomena could
be related to the impact in the global spontaneous emission of the two contributions in Eq. (6.13), namely that of the homogeneous part \(|c_2(0,t)|^2\) and the one originating from noise operators in the time integral in the last line. In the inset of Fig. 6.6 we have depicted the ratio of the homogenous solution with the global emission at \(k = 0^+\), calculated without \(\langle \hat{P}_k^\dagger \hat{P}_k \rangle_{PL}\). Rather surprisingly when increasing the pump intensity the two contributions (homogeneous and particular) continue to have comparable contributions, hence for a proper description of the spontaneous parametric emission they must be both included.

In Fig. 6.7, are shown 6 snapshots at different times of the 2 dimensional mapping in \(k\)-space of the polariton occupation for the structure of Ref. [14] consisting of a 25 nm GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As single quantum well placed in the center of a \(\lambda\) cavity with AlAs/Al\(_{0.15}\)Ga\(_{0.85}\)As Bragg reflectors. The pump is set above the magic angle, i.e. \(k_p = (1.73 \mu m^{-1}, 0.)\). The different figure colours are normalized so that to enhance all the features in each graphs, thus they are to be considered to give the qualitative temporal dynamics. As can be clearly seen in the figures the build-up of the eight-shaped curve for the resonant parametric processes takes place around \(t = 6\) ps, but only from \(t = 9\) ps start to overcome the incoherent dynamics. Being a pulsed excitation the dynamics reaches its maximum around \(t = 16\) ps and decreases afterwards. We do not have considered, only for the sake of presentation, an incoherent noisy background due, as instance, to the photodetector noise, however in our approach would be very easy to include it in future simulation of real experiments.

Fig. 6.8(b) shows the time integrated mapping of the dynamics of Fig. 6.7, whereas Fig. 6.8(a) refers to the same structure, excited with the same pulse shape, but resonant exactly to the magic angle \(k_m\). We can see that on the contrary to the case of an excitation at \(k\)’s above the magic angle when the sample is excited at \(k_m\) the eight-shaped curve, theoretically predicted as an energies plus wave vectors conservation, starts to be unclear. Only at higher pumping angles the phase-space
Figure 6.7. (color online). Snapshots at different times of the two dimensional dynamics for the structure of Ref. [14] consisting of a 25 nm GaAs/Al$_{0.3}$Ga$_{0.7}$As single quantum well placed in the center of a $\lambda$ cavity with AlAs/Al$_{0.15}$Ga$_{0.85}$As Bragg reflectors. The pump is set above the magic angle, i.e. $k_p = (1.73\mu m^{-1},0.)$.

for the allowed resonant parametric processes has a clear eight-shaped structure. We impute this behaviour to the difference between the fulfilment of a set of coupled identities and the spanning of $k$-space by polaritons during their real dynamics which does depend on various competing contributions. Also in recent experiments
an analogous fact has been reported and we consider our result as its possible explanation.

The calculated time-integrated spectra of the outgoing light at $k = (0,0)$ obtained at six different pump intensities for an excitation at the magic angle $k_m$ are shown in Fig. 6.9. It can be easily noticed a threshold around $I^L = 20 I_0$ in perfect agreement with the results in Fig. 6.5 and with Ref. [14]. For intensities lower than the threshold, the signal in $k_x = 0$ (with the corresponding idler in $k_x = 2k_m$)
Figure 6.9. Calculated time-integrated spectrum of the outgoing light at $k = (0,0)$ normalized with respect to the pump seed obtained at six different pump intensities for an excitation at the magic angle $k_m$. It can be easily noticed a threshold around $I_L = 20 I_0$ in perfect agreement with the results in Fig. 6.5 and with Ref. [14]. For intensities lower than the threshold, the signal in $k_x = 0$ (with the corresponding idler in $k_x = 2k_m$) shows a quite large nearly Lorentian shape, as soon as the threshold is passed over the spectrum start to increase super-linearly meanwhile the linewidth decreases witnessing the parametric emission build-up. In the inset the normalized spectra at increasing pump powers (indicated by the arrow direction) are depicted evidencing even better the linewidth narrowing. We notice also some spurious queues due to (calculated) asymmetric signal/idler damping values.
a narrow linewidth beyond the mentioned threshold.
Part III

Coherent and incoherent optical properties in the weak-coupling case.

Application to near-field optical spectroscopy
Chapter 7

Introduction

Investigation of the ultrafast dynamics has become in recent years a strategic field both in research and from a technological point of view. Recent developments in ultrafast laser physics and technology now allow to study the very initial interaction processes of non-equilibrium carriers in a semiconductor [26, 27] directly related to the microscopic details of the coupling mechanisms. As a consequence time-resolved laser spectroscopy has become an essential precious tool in modern semiconductor physics. Linear optical spectroscopy of semiconductor has provided invaluable information about electronic band structures, phonons, plasmons, single-particle spectra and/or defects. Though impressive contributions, however, information on the details of the interaction processes among the elementary excitations are often much more difficult. On the contrary, an optical excitation has the ability to generate non-equilibrium carrier and exciton distributions and time-resolved spectroscopy provides the best mean of determining the temporal evolution of such distribution functions. Indeed, ultrashort optical pulses may generate coherent superpositions of states and the dynamics of such phase-related quantities can be analyzed. Furthermore when combined with spatial imaging techniques and/or specific low dimensional structures, ultrafast optical spectroscopy becomes a very powerful tool.
for investigating a wide variety of phenomena related to relaxation and transport
dynamics in semiconductors [27]. Typical time scales for the various coherent and
incoherent microscopic mechanisms whose interplay and competition constitute the
very non-trivial problem in dealing with such phenomena both theoretically and
experimentally are in the range of pico- and femtoseconds, therefore the dynamics
generated is generally called \textit{ultrafast}. The aim of an ultrafast optical experiment
is to provide information on the details of the underlying temporal evolution which,
in turn, gives insight into the fundamental processes governing the microscopic car-
rier dynamics. A typical scenario for the dynamics of distribution functions is easy
explained: the laser pulse with a given photon energy and a certain spectral width
determined by its duration creates electron-hole pairs in a more or less localized
region in \textbf{k}-space. This initial distribution then relaxes due to the presence of scat-
tering processes, describing the various interactions the system of interest has with
all the other external degrees of freedom such as lattice vibration, before being
re-emitted by radiative recombination. These thermalization and relaxation pro-
cesses have been studied in great detail in the past two decades both experimentally
and theoretically in bulk semiconductor materials as well as in a wide variety of
heterostructures. The most commonly used experimental techniques have been lu-
minescence (where the photons created by the radiative recombination of electron
and holes are detected) and pump-probe (PP) measurements (where, in turn, the
change in the absorption or in reflection of a probe beam caused by the prior ex-
citation of the electron-hole pairs by a pump beam is observed), see e.g. [41, 46]
and references therein. Besides PP and luminescence measurements, there are other
techniques which rely completely on the phase coherence in the carrier system, pro-
viding a direct information on the dynamics of coherent interband and intraband
polarizations such as, for instance, four-wave-mixing (FWM) experiments.
Excitons (or interband polarizations) are sensitive two the reduced dimensionality of the semiconductor structure. Nanostructured heterostructures may confine the carrier wave function in one or more dimensions, by constructing such ”artificial structure”, it is possible to modify and control the optical and transport properties of the system. In a QW the wave function of the first excited electronic states (those closer the band edge and thus participating in the exciton optical formation) can be confined in the growth direction whereas they are of Bloch type in the in-plane orthogonal directions. The electron-hole confinement enhances the interaction with the radiation and generically alters the incoherent scattering features. As a consequence optical as well as transport physics are subjected to many modifications due to the reduced dimensionality. For instance, the confinement determines a lacking of translational symmetry in the growth direction (for convention the z-axis) and only the orthogonal components of the wave vector, \( k^\parallel \), is conserved. It means that an exciton with a given \( k^\parallel \) can interact with those light modes of the same \( k^\parallel \), but with generic \( k_z \), thus excitons in QW show a radiative decay analogous to the spontaneous emission of the atomic electronic levels on the contrary to the bulk case where the full \( k \) conservation makes the radiative decay to be absent [43].

Near-field optical microscopy and spectroscopy, which uses optical interaction in the visible or near-infrared range has demonstrated its ability to image optical fields and surface structures at a sub-wavelength scale. In particular, the ability of this kind of optical microscopy and spectroscopy to identify the individual quantum constituents of semiconductor quantum structures has been widely demonstrated [87, 88, 89]. In the last years, scanning near-field optical microscopy (SNOM) and spectroscopy opened the possibility for studying optical phenomena with resolution well below the diffraction limit for quantum systems in a complex solid state environment. SNOM combines the advantages of nanometric resolution of scanning-probe microscopy with the unique possibility of characterizing quantum systems offered
by optical spectroscopy. A rich area of physics involving spatially resolved quantum systems in a complex solid state environment provided direct information on the spatial and energy distribution of light emitting nanometric centers of semiconductor quantum structures [87, 88, 89, 31, 91, 92]. A recent work by Runge and Lineau [98] analyzes the interpretation of near-field images of semiconductor nanostructures; they conclude that direct imaging of the exciton density is only possible in collection mode experiments with nonresonant excitation in the high-resolution limit. For other geometries and for biexcitonic states, the images reflect not only the size and shape of the wave function and the spatial resolution of the near-field probe but also in particular the inherent optical nonlinearity of the imaging process. 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 [42]. The mean-field (MF) is a cornerstone of the ultrafast dynamics at the semiconductor band-edge [41]. However, this level of the theory does not provide for a microscopic description of key effects in semiconductor quantum optics as Coulomb scattering and two-pair correlations. In studying optical processes in semiconductor systems, lots of things can be learned form ensembles of noninteracting atomic transition interacting with the radiation field; the equations arisen in this context are widely known as optical Bloch equations. However, the proximity of the atoms in forming a solid and the strong Coulomb interaction between electrons give phenomena that cannot be explained within such a simplified model. In [83] a generalization of the previous theory was presented, it described the dynamics of semiconductor electrons under the influence of driving laser fields including Coulomb correlation between electrons, they were called Semiconductor Bloch Equations (SBE). Anyway, they display the structure of an open hierarchy of equations, thus bearing the problem of finding
an appropriate truncation procedure. In 1994 and 1995 [60, 59] a new idea was presented in the literature, the so-called Dynamics Controlled Truncation Scheme (DCTS). It provides a (widely adopted) starting point for the microscopic theory of the light-matter interaction effects beyond mean-field [24] supplying a consistent and precise way to stop the infinite hierarchy of higher-order correlations which always appears in the microscopic approaches of manybody interacting systems. In 1996 the DCTS was extended in order to include in the description the quantization of the electromagnetic field [20]. Within this approach, once fixed the perturbative order in the excitation strength – this way deciding the nonlinear optical processes sought – the Coulomb correlation contributing up to the given order is included exactly. Indeed it can be proved that the third order nonlinear response depends only on the one eh-pair (excitons) and the two eh-pair (biexcitons or four particles aggregates) subspaces, hence it is clear that up to the third order optical nonlinear response is intimately related to two- and four-particle quantum coherences.

In this chapter we follow the near-field theoretical model of Ref. [32], though in a more general framework. We calculate and introduce the time equation for the most generic quantity to be addressed in order to study the optical response under coherent excitation of a generic QW in the weak-coupling case and we will be able to give at any order of the coherent exciting field the system of first order differential equations to be solved numerically in order to gather the output optical field witnessing the nonlinear dynamics inside the heterostructure. The first case-study we shall analyzed will be near-field spectroscopy of QW with interface disorder, i.e. naturally occurring quantum dots. In order to do this we recover the equation of Ref. [32] simply as the linear order of ours. The numerical result here presented are those we published in [84].
Chapter 8

Near-field optical spectroscopy: photoluminescence at any exciting order

In the previous part the so-called *coherent limit* has been extensively used when we described the biexciton contribution as a PSF, a MF and an exciton-exciton interaction terms in Chapter 4, that is two say it has been assumed that the coherent pumping generate *coherent* parametric processes. This way one is able to describe the biexcitonic contributions effectively as exciton correlations. For the sake of clarity, for a generic operators as the projection operator $\hat{X}_{N\alpha,M\beta}$ build up through the interaction many body Hamiltonian of the photo-excited system, we may think without loss of generality that

$$\langle \hat{X}_{N\alpha,M\beta} \rangle = \langle \hat{X}_{N\alpha,M\beta}^{\text{coh}} \rangle + \langle \hat{X}_{N\alpha,M\beta}^{\text{inc}} \rangle.$$  \hspace{1cm} (8.1)

The coherent limit comprised to neglecting the latter addendum considering it has negligible contribution in a mostly coherent optical process. We tried not to use
this approximation and we succeeded in constructing a microscopic model for envi-
ronment noise and decoherence in resonant emission form QW in the weak coupling
regime.

The full Hamiltonian describing the system is:

\[ \hat{H} = \hat{H}_c + \hat{H}_0^{\text{phon}} + \hat{H}_c^{\text{light}} + \hat{H}_c^{\text{sem}} + \hat{H}_c^{\text{ep}}, \]  
\[ \text{(8.2)} \]

where

\[ \hat{H}_c = \hbar \sum_{N\alpha} \omega_{N\alpha} \hat{X}_{N\alpha} \hat{X}_{N\alpha}; \] \[ \hat{H}_0^{\text{phon}} = \hbar \sum_{\mathbf{q}} \omega_{\mathbf{q}} \hat{b}_\mathbf{q} \hat{b}_\mathbf{q}. \]  
\[ \text{(8.3)} \]

The acoustic phonon interaction Hamiltonian reads:

\[ \hat{H}_c^{\text{phon}} = \sum_{N\zeta} \sum_{\mathbf{q}} t^q_{N\zeta} \hat{X}_{N\zeta} \hat{X}_{N\zeta} (\hat{b}_\mathbf{q} + \hat{b}_\mathbf{q}^\dagger), \]  
\[ \text{(8.4)} \]

where \( \zeta, \eta \) are the proper quantum number spanning the associated N-pair state
space (e.g. if \( N = 1 \) \( \zeta = (n_{\zeta, \sigma_{\zeta}}) \)). In the Hamiltonian above we have include all the
other ingredients inside \( t \):

\[ t^q_{N\zeta} = \sum_{\mathbf{k}} \left( \frac{\hbar |\mathbf{q}|}{2\rho v_s N} \right)^{1/2} \left( \langle N\zeta | D_c \hat{c}_{\mathbf{k+q}}^\dagger \hat{c}_{\mathbf{k}} + D_v \hat{d}_{\mathbf{k+q}}^\dagger \hat{d}_{\mathbf{k}} | N\eta \rangle \right). \]  
\[ \text{(8.5)} \]

Here there are the deformation potential coupling constants for the conduction and
valence bands, the mass density and the velocity of sound for the longitudinal acous-
tic phonons [49].

As for the electromagnetic light field we treat it, at least as a first step, as divided
in two different parts: a semiclassical coherent and a fully quantum incoherent ones.
In the usual rotating wave approximation it can be written as

\[ \hat{H}^{\text{int}} = -\int d^3r \hat{E}(\mathbf{r}) \hat{P}^\dagger(\mathbf{r}) + H.c. \]  
\[ \text{(8.6)} \]

We separate the field operator \( \hat{E}(\mathbf{r}) \) into a fluctuating part \( \hat{E}_{\mathbf{k}} \) (the one determining
the spontaneous emission), that can be expanded in terms of annihilation photon
operators and has the properties [85]:

\[ [ \hat{E}_k, \hat{E}^\dagger_{k'} ] = g_k \delta_{k,k'}, \langle \hat{E}^\dagger_k \hat{E}_{k'} \rangle = n_k \delta_{k,k'}, \]  (8.7)

and into a classical contribution \( E_I (r) \), this way we shall describe the (possibly inhomogeneous) exciting electric field \( (E_{in}) \) as well as the signal mode delivered by the tip in a SNOM setup \( (E_{out}) \):

\[ \hat{E} (r) = \sum_k \hat{E}_k e^{ik \cdot r} + E_I (r) . \]  (8.8)

In this way, we are assuming that the presence of the illuminating tip does not alter the radiative decay rates.

The interband polarization density operator can be expressed in terms of exciton operators as

\[ \hat{P} (r) = -\mu_{cv} \sum_\alpha f (z) \phi_{1s} (0) \psi_{\alpha}^* (R) \hat{B}_\alpha, \]  (8.9)

so, our light interaction Hamiltonian becomes

\[ \hat{H}_{int} = \int dzd^2 R \left( \sum_k \hat{E}_k e^{ik \cdot r} + E_I (r) \right) \mu_{cv} \sum_\alpha f (z) \phi_{1s} (0) \psi_{\alpha}^* (R) \hat{B}_\alpha^\dagger \\
+ H.c., \]  (8.10)

a sum of a quantum and a semiclassical contribution:

\[ \hat{H}^q_{int} = \hat{H}^q_{LM} + \hat{H}^{sem}_{LM} . \]  (8.11)

The first one can be written as

\[ \hat{H}^q_{LM} = \sum_{\alpha k} \hat{E}_k M_\alpha^\ast (k) \hat{B}_\alpha^\dagger + H.c., \]  (8.12)

where

\[ M_\alpha (k) = \mu_{cv} \phi_{1s} (0) \int dzd^2 R f (z) e^{-ik \cdot r} \psi_{\alpha} (R) , \]  (8.13)
and the latter one can be synthetically written as

\[ \hat{\mathcal{H}}_{\text{LM}}^{\text{sem}} = \sum_{\alpha} E_{I\alpha} \hat{B}_{\alpha}^\dagger + H.c., \]  

(8.14)

where

\[ \hat{E}_{I} (R) = \int dz E_{I} (r) f(z), \]  

(8.15)

\[ O_{I\alpha} = \int d^2R \psi_{\alpha}^* (R) \hat{E}_{I} (R), \]  

(8.16)

\[ E_{I\alpha} = \mu_{c\alpha} \phi_{1s} (0) O_{I\alpha}. \]  

(8.17)

Setting as usual our notation of the generic Hubbard operator \( \hat{X}_{N\alpha;M\beta} \), that is to say a generalized projection operator onto exact eigenstates of the Npair e-h Hamiltonian we have the exact identity which states that the bases is complete:

\[ I = \sum_{N\alpha} \hat{X}_{N\alpha;N\alpha}, \]  

(8.18)

to express the exciton operator as

\[ \hat{B}_{\alpha} = \hat{X}_{0;1\alpha} + \sum_{N \geq 1} \sum_{\eta, \beta} \langle N\eta | \hat{B}_{\alpha} | (N+1)\beta \rangle \hat{X}_{N\eta;(N+1)\beta}. \]  

(8.19)

From the form of the interaction and of the pump Hamiltonian we describe the optical response on a coherent input light field within the DCTS. As already written in detail in Chapter 2.2 the theorem states that the expectation values of these operators can be expressed as a power series in the input field.

\[ \langle \hat{X}_{N;\alpha;M,\beta} \rangle = \sum_{i=0}^{i_0} \langle \hat{X}_{N;\alpha;M,\beta} \rangle^{(N+M+2i)} + O(c^{(N+M+2i+2)}). \]  

(8.20)

The state of the quantum system is defined by a density operator and a master equation describing its time evolution. The full analytic solution of such an equation is not known even for the much simpler two level atoms single mode cavity system. Instead of solving the master equation, we consider the equation of motion for the expectation values of system operators of interest, up to a given order.
8.1 Dark states photoluminescence in naturally occurring quantum dots: far- and near-field spectroscopy

The optical spectra of semiconductors provide a rich source of information on their electronic properties. In a typical luminescence experiment one excites initially a nonequilibrium distribution of electron-hole (e-h) pairs in a semiconductor. In most cases the electrons and holes will thermalize among themselves and reach quasithermal equilibrium in a time short compared to the time it takes for electrons and holes to recombine [86]. In the final step the e-h pairs recombine radiatively, producing spontaneous emission. Understanding how electron-hole pairs distribute among the available energy levels of semiconductor nanostructures after optical excitation or current injection is crucial for the development of novel quantum devices [99]. One tool frequently used to explore the energy distribution of electron-hole pairs is photoluminescence spectroscopy (PL). It provides a direct measurement of the optical density of states times the excitonic population density as a function of energy. Detailed simulations in Refs. [100, 93] have clarified many aspects of the nonequilibrium dynamics determining the distribution of excitonic populations among the available energy levels in quantum wells (QWs) with interface roughness. Already at temperatures around 40-50 K the individual occupation of states is close to the Boltzmann distribution since all states can frequently emit and absorb phonons to reach equilibrium before radiative emission. At lower temperature more localized states displaying smaller phonon scattering rates cannot equilibrate and their occupations deviate from the Boltzmann distribution, displaying a reduced energy dependence. Far-field PL spectroscopy provide average measurements over a distribution of different emission sites. The opportunity for important insight is often lost by the inability to resolve finer details within this distribution. Scanning
near-field optical microscopy (SNOM) combines the advantages of nanometric resolution of scanning-probe microscopy with the unique possibility of characterizing quantum systems offered by optical spectroscopy. The SNOM ability to resolve the individual quantum constituents of semiconductor nanostructures has been widely demonstrated [87, 101, 102]. If the spatial near-field resolution falls below the extension of confined quantum systems, spatially resolved PL maps out the spatial probability distribution of the wave function times the corresponding level occupation [32, 98].

Among the most investigated quantum structures are quantum dot (QD) systems formed naturally by interface fluctuations in narrow quantum wells [90]. QD arrays have distributions in size and shape that lead to inhomogeneous spectral linewidth and averaged data. To avoid this problem, these systems are mainly probed by means of techniques of high resolution spatially resolved spectroscopy and microscopy. In particular, near-field optical measurements based on spatially-resolved photoluminescence (PL) can detect the optical characteristics of individual quantum dots (QDs) among e.g. a high-density ensemble of naturally formed QDs [90], whereas usual far-field methods provide only ensemble-averaged properties. Detailed simulations of Zimmermann et al. [93, 85] have clarified many aspects of the intrigued non-equilibrium dynamics giving rise to PL spectra in these quantum structures. Theoretical simulations of near-field imaging spectroscopy of semiconductor quantum structures have focused their attention on calculations of local absorption [94, 95, 96, 97]. The matrix elements governing the light-matter coupling are a convolution of the quantum states with the exciting electromagnetic field. This convolution implies that exciting a direct gap bulk semiconductor with a light field of a given wavevector resonant with the energy gap results in exciting excitons (i.e. bound state interband optical transitions) within the same wavevector. Succeeding in confining the optical excitation to a very small volume below the diffraction
limit, implies the presence of optical fields with high lateral spatial frequencies, determining coupling matrix elements that can differ significantly from far-field ones [32, 94, 95, 96, 97, 103]. The most striking manifestation of these effects is the breaking of the usual optical selection rules and the possibility to excite dark states whose optical excitation is forbidden by symmetry in the far field. Spatial maps of dark states in semiconductor nanostructures were simulated for high-resolution SNOM in absorption and two-photon experiments [94, 104, 105, 106]. Moreover, dark states are not able to emit light in the far-field, for they generate only fields with high in-plane wavevectors corresponding to evanescent waves. Hence their lifetimes at low temperatures result significantly increased with respect to their bright counterparts. Recently, a microscopic analysis of spatially resolved PL and photoluminescence excitation (PLE) spectroscopy in semiconductor quantum structures [32] was presented. In particular, in Ref. [32] numerical results are shown that clarify the impact of the near-field optical setup on the obtained images and resolutions. The microscopic theory of spatially resolved PL in quantum structures here presented and exploited includes both light quantization (essential to describe radiative recombination) and phonon scattering. We faced with a strictly non-equilibrium problem arising from both radiative recombination (preventing full thermalization) and from the local nature of the excitation source (in the illumination-mode setup).
8.1.1 Disorder and Center of mass motion

Structural disorder on mesoscopic length scale is an unavoidable feature of semiconductor nanostructures. In addition to the impurities, lattice imperfections, etc., which are also present in homogeneous bulk crystals, semiconductor quantum structures additionally exhibit interface imperfections which can involve different components with different lateral length scales. Despite the progress in atomic-layer epitaxy, the fabricated interfaces are far from being perfect and exhibit monolayer islands and terraces with lateral extent depending on the growth conditions. Furthermore, atomic interdiffusion and fluctuations of alloys composition determine potential fluctuations which influence the behaviour of photexcited carriers [107, 108].

In this context, the exciton Schrödinger equation for a two-band model in effective mass approximation can be written as [85]

\[
\left( -\frac{\hbar^2}{2m_e^*} \nabla^2_{r_e} - \frac{\hbar^2}{2m_h^*} \nabla^2_{r_h} - \frac{e^2/\epsilon_0}{|r_e - r_h|} + W_e(r_e) + W_h(r_h) - \epsilon_\alpha \right) \Psi_\alpha(r_e,r_h) = 0 ,
\]

(8.21)

where the standard kinetic terms for the electron and the hole (with effective masses \(m_e^*, m_h^*\) and positions \(r_e, r_h\) respectively) and the Coulomb attraction term appear. We have introduced the confinement potentials \(W_e(r_e)\) and \(W_h(r_h)\) which describe the spatial variation of the local band edges. If interface roughness dominates the disorder, it depends on the band edge difference between barriers and well material and on the local well width [85].

If the exciton binding energy is small compared to the energetic sublevel distance, but also well above the disorder-induced broadening, only the lowest bound state \(1s\) at the fundamental sublevel transition has to be considered and the total wave function can be factorized into

\[
\Psi_\alpha(r_e,r_h) = u_e(z_e) u_h(z_h) \phi_{1s}(\rho) \psi_\alpha(R) ,
\]

(8.22)
where we have introduced the 2D center of mass coordinate
\[
\mathbf{R} = \frac{m_e^* \mathbf{r}_e^\parallel + m_h^* \mathbf{r}_h^\parallel}{M}, \quad \rho = \mathbf{r}_e^\parallel - \mathbf{r}_h^\parallel,
\] (8.23)
with the exciton kinetic mass
\[
M = m_e^* + m_h^*
\] (8.24)
and \( \mathbf{r}_e^\parallel, \mathbf{r}_h^\parallel \) the projection vectors of \( \mathbf{r}_e, \mathbf{r}_h \) in the plane of the quantum well. Both the confinement wave functions \( u_e(z_e), u_h(z_h) \) and the relative wave function \( \phi_{1s}(\rho) \) obey Schrödinger equations of the QW structure with average thickness \( L_z \) (as a convention \( z \) has been taken as the coordinate along the growth direction) [109].

Finally, one is left with the COM equation
\[
\left( -\frac{\hbar^2}{2M} \nabla_{\mathbf{R}}^2 + V(\mathbf{R}) \right) \psi_\alpha(\mathbf{R}) = \epsilon_\alpha \psi_\alpha(\mathbf{R}),
\] (8.25)
where \( V(\mathbf{R}) \) is the random COM potential resulting from well-width fluctuations. The normalization is taken over the in-plane area \( A \), with orthogonality relation
\[
\int_A d\mathbf{R} \psi_\alpha(\mathbf{R}) \psi_\beta(\mathbf{R}) = \delta_{\alpha,\beta}.
\] (8.26)

The treatment above can be greatly expanded in order to consider \( N \) pair aggregate and not only exciton (i.e. 1 pair aggregate), see Appendix (B).
8.1.2 Damping terms in Markov approximation

For the narrow energy window around the 1s exciton, it suffices to construct an exciton Hamiltonian from the underlying electron-hole picture. Traditionally, this is done by defining creation and annihilation operators of excitons 1S in plane-wave COM states. Then, the disorder enters as an elastic scattering process, commonly treated in second Born approximation. Our emphasis, however, is on a proper description of disorder effects. Therefore, we prefer to work in the representation of disorder eigenstates and assign to each of the states $\alpha$ an exciton creation operator $\hat{B}_\alpha^\dagger$. As a consequence the exciton wave vector $\mathbf{k}$ is no more a good quantum number and the acoustic phonon interaction Hamiltonian changes, though slightly, its form:

$$
\hat{H}_{ep} = \sum_{N\alpha\beta} \Gamma_{N,\alpha\beta}^q \left( \hat{b}_q^\dagger + \hat{b}_{-q} \right) \hat{X}_{N\alpha;N\beta}.
$$

(8.27)

The damping, within Markov approximation, are given in details in Appendix A for the non disordered case. However with slight changes the same steps can be followed also here. Thus we referred to the Appendix A for the lengthy calculation which bring us to obtaining:

acoustic phonons

$$
\frac{d}{dt} \langle \hat{X}_{N\alpha;M\beta} \rangle_{|_{\hat{H}_{ep}}} = -\frac{1}{2} \left( \Gamma_N^\alpha + \Gamma_M^\beta \right) \langle \hat{X}_{N\alpha;M\beta} \rangle + 
+ \frac{1}{2} \delta_{M,N} \delta_{\alpha,\beta} \sum_{\zeta} \left( W_{\alpha,\zeta}^{(N)} + W_{\beta,\zeta}^{(M)} \right) \langle \hat{X}_{N\zeta;N\zeta} \rangle,
$$

(8.28)
with the usual scattering rates.

\[
W_{\eta,\alpha}(N)^\pm = \frac{2\pi}{\hbar} \sum_q |t_{N\eta,\alpha}^q|^2 \delta(\hbar\omega_{N\eta} - \hbar\omega_{N\alpha} \pm \epsilon_q^R) (n_q^R + \theta(\pm))
\]

\[
W_{\eta,\alpha}(N) = \sum_\pm W_{\eta,\alpha}(N)^\pm
\]

\[
\Gamma_{\alpha}^N = \sum_\zeta W_{\eta,\alpha}(N),
\tag{8.29}
\]

as usual, we have adopted the \textit{matrix convention} that is to say in \(W_{\eta,\alpha}(N)\), for instance, we are considering \(\alpha\) as the initial state and \(\eta\) the final state after the scattering event, \(N\) refers to the number of e-h pairs into play.

**radiative scattering**

\[
\frac{d}{dt} \langle \hat{X}_{N\alpha;M\beta}\rangle |_{\rho_{N,M}^{\beta}} = -\frac{1}{2} (\Gamma_{N\alpha}^l + \Gamma_{M\beta}^l) \langle \hat{X}_{N\alpha;M\beta}\rangle +
\]

\[
+ \delta_{N,M} \delta_{\alpha,\beta} \sum_\rho \left[ W_{N\alpha,(N+1)\rho}^{(-)l} \langle \hat{X}_{(N+1)\rho;(N+1)\rho}\rangle +
\right.
\]

\[
\left. + W_{N\alpha,(N-1)\rho}^{(+)l} \langle \hat{X}_{(N-1)\rho;(N-1)\rho}\rangle \right]
, \tag{8.30}
\]

where

\[
W_{RF;P_i}^{(+)} = \frac{2\pi}{\hbar} \sum_{k\eta} |M_{\eta}(k)|^2 \left[ n_k^l \right] \delta(\hbar\omega_R - \hbar\omega_{P_i} - \hbar\omega_k^l) |Rf | \hat{B}_\eta | P_i\rangle|^2
\]

\[
W_{RF;P_i}^{(-)} = \frac{2\pi}{\hbar} \sum_{k\eta} |M_{\eta}(k)|^2 \left[ n_k^l + g_k \right] \delta(\hbar\omega_R - \hbar\omega_{P_i} + \hbar\omega_k^l) |Rf | \hat{B}_\eta | P_i\rangle|^2
\]

\[
\Gamma_{Qq}^l = \sum_\rho \rho_{Qq}^{\pm l} W_{Q\pm l;Qq}^{\pm l}
, \tag{8.31}
\]

The generic equation of motion then reads:
\[
\frac{d}{dt} \langle \hat{X}_{N\alpha,M\beta} \rangle = -\frac{1}{2} \left[ \left( \Gamma^N_{\alpha} + \Gamma^M_{\beta} \right) + \left( \Gamma^l_{N\alpha} + \Gamma^l_{M\beta} \right) \right] \langle \hat{X}_{N\alpha;M\beta} \rangle + \\
+ \delta_{M,N} \delta_{\alpha,\beta} \left[ \sum_\zeta W^{(N)}_{\alpha,\zeta} \langle \hat{X}_{N\zeta;N\zeta} \rangle + \sum_\rho \left( W^{(-)l}_{N\alpha,(N+1)\rho} \langle \hat{X}_{(N+1)\rho;(N+1)\rho} \rangle + \right. \right. \\
+ \left. \left. W^{(+)}l_{N\alpha,(N-1)\rho} \langle \hat{X}_{(N-1)\rho;(N-1)\rho} \rangle \right) \right] + \\
+ \sum_{\pi\rho} \frac{i}{\hbar} \epsilon_{I\pi} \left( \langle M\beta | \hat{B}_{\pi}^\dagger | (M-1)\rho \rangle \langle \hat{X}_{N\alpha;(M-1)\rho} \rangle - \langle (N+1)\rho | \hat{B}_{\pi}^\dagger | N\alpha \rangle \langle \hat{X}_{(N+1)\rho;M\beta} \rangle + \right. \\
+ \left. \frac{i}{\hbar} \epsilon_{I\pi}^* \left( \langle M\beta | \hat{B}_{\pi} | (M+1)\rho \rangle \langle \hat{X}_{N\alpha;(M+1)\rho} \rangle - \langle (N-1)\rho | \hat{B}_{\pi} | N\alpha \rangle \langle \hat{X}_{(N-1)\rho;M\beta} \rangle \right) \right), \\
\text{(8.32)}
\]

where \( \epsilon_{I\alpha} \) has been defined in (8.17). If the disorder induced broadening is small compared to the exciton binding energy, only the lowest bound state 1s at the fundamental sublevel transition has to be considered and the exciton wave function can be factorized. In this situation we shall consider only the lowest order contribution, i.e. that of 1-pair aggregate only. For notation simplicity we will not write the order we are moving into, on the contrary from now on it will be always the lowest level (i.e. 1 for 1-body operator and 2 for two-body operator such as exciton density).

The kinetic equation for diagonal terms of the exciton density matrix \( N_{\alpha\beta} = \langle \hat{X}_{1\alpha;1\beta} \rangle \) can be derived starting from the Heisenberg equation of motion for the exciton operators under the influence of \( H \). We define \( P_\alpha = \langle \hat{X}_{1\alpha,0} \rangle, \Gamma_\alpha = \Gamma^1_{\alpha} + \Gamma^l_{1\alpha} \). According to the perturbative level \( W^{(-)l} \) enters as an higher order, thus not to be included, \( W^{(+)}l \) is proportional to \( n^l_k \) considered negligible in order to include in the description only the spontaneous radiative decay. The total radiative out-scattering
thus reads

\[ \Gamma_{l1} = \sum_{\rho} W_{2p,1\alpha}^{(+)} + W_{0,1\alpha}^{(-)} = W_{0,1\alpha}^{(-)} = \]

\[ = \frac{2\pi}{\hbar} \sum_{k} |M_{\alpha}\rangle |n_{k}^{+} + g_{k}\rangle \delta(-\hbar \omega_{1\alpha} + \hbar \omega_{k})\langle 0 \mid \hat{B}_{\alpha} \mid 1\rangle|^{2} = \]

\[ = \frac{2\pi}{\hbar} \sum_{k} |M_{\alpha}\rangle |g_{k}\rangle \delta(\hbar \omega_{k} - \hbar \omega_{1\alpha}) , \quad (8.33) \]

We obtain

\[ \partial_t N_{\alpha\beta} (t) = \left( i(\omega_{\alpha} - \omega_{\beta}) - \frac{1}{2}(\Gamma_{\alpha} + \Gamma_{\beta}) \right) N_{\alpha\beta} (t) + \delta_{\alpha\beta} \sum_{\rho} W_{\alpha,\rho}^{(1)} N_{\rho\rho} (t) + \]

\[ + \frac{i}{\hbar} \mathcal{E}_{I\alpha}^* P_{\beta} (t) - \frac{i}{\hbar} \mathcal{E}_{I\beta} P_{\alpha} (t) \quad (8.34) \]

\[ \partial_t P_{\alpha} (t) = \left( i\omega_{\alpha} - \frac{1}{2}\Gamma_{\alpha} \right) P_{\alpha} (t) + \frac{i}{\hbar} \mathcal{E}_{I\alpha} . \quad (8.35) \]

The polarization equation can be immediately solved and gives

\[ P_{\alpha} (t) = \int_{-\infty}^{t} dt' e^{(i\omega_{\alpha} - \frac{1}{2}\Gamma_{\alpha})(t-t')} \frac{i}{\hbar} \mathcal{E}_{I\alpha} . \quad (8.36) \]

Instead, Eq. (8.34) is solved by the factorized ansatz

\[ N_{\alpha\beta} (t) = P_{\alpha} (t) P_{\beta}^* (t) , \alpha \neq \beta , \quad (8.37) \]

for the nondiagonal density matrix \( N_{\alpha\beta} \) with \( \alpha \neq \beta \), whereas the kinetic equation of the diagonal density \( N_{\alpha\alpha} (t) \) needs to be solved numerically

\[ \partial_t N_{\alpha\alpha} (t) = S_{I\alpha} (t) + \sum_{\rho} W_{\alpha,\rho}^{(1)} N_{\rho\rho} (t) - \Gamma_{\alpha} N_{\alpha\alpha} (t) , \quad (8.38) \]

with the source term

\[ S_{I\alpha} (t) = Im \frac{2}{\hbar} P_{\alpha} (t) \mathcal{E}_{I\alpha} . \quad (8.39) \]

For stationary excitation, the source term becomes time-independent and can be written as

\[ S_{I\alpha} = \frac{2}{\hbar^2} |\mathcal{E}_{I\alpha}|^2 \frac{\Gamma_{\alpha}/2}{(\omega - \omega_{\alpha})^2 + \Gamma_{\alpha}^2/4} . \quad (8.40) \]
8.1.3 Near-field model

The positive frequency components of the operator describing the signal that can be detected by a general near-field setup can be expressed as [110]

$$\hat{S}_t^+ = \hat{A}_{bg}^+ + \hat{S}^+, \quad (8.41)$$

where $\hat{A}_{bg}^+$ is the elastic background signal (largely uniform along the $x-y$ plane) proportional to the input electric-field operator, and $\hat{S}^+$ is related to the sample polarization density operator $\hat{P}^\dagger(\mathbf{r})$:

$$\hat{S}^+ = \mathcal{A} \int d\mathbf{r} \hat{P}^\dagger(\mathbf{r}) \cdot \mathbf{E}_{out}(\mathbf{r}), \quad (8.42)$$

where $\mathcal{A}$ is a complex constant and $\mathbf{E}_{out}(\mathbf{r})$ is the signal mode delivered by the tip. Photoluminescence can be defined as the incoherent part of the emitted light intensity [111]. The PL that can be measured by a photodetector after the collection setup (broadband detection) is proportional to $I = \langle \hat{S}^- , \hat{S}^+ \rangle$, with $\langle \hat{A} , \hat{B} \rangle \equiv \langle \hat{A} \hat{B} \rangle - \langle \hat{A} \rangle \langle \hat{B} \rangle$, thus we may define

$$I_{PL} = I - I_{coh} = \langle \hat{S}^- , \hat{S}^+ \rangle = \langle \hat{S}^- (\tau) \hat{S}^+(\tau) \rangle - |\langle \hat{S}^\dagger(\tau) \rangle|^2. \quad (8.43)$$

Analogously, the steady-state spectrum of incoherent light emitted by the semiconductor quantum structure and detected by the SNOM setup can be expressed as

$$I_{PL}(\omega_{out}) = \frac{1}{\pi} \mathcal{R} \int_0^\infty d\tau \langle \hat{S}^- (0) , \hat{S}^+(\tau) \rangle e^{i\omega_{out}\tau}. \quad (8.44)$$

Once the exciton densities have been derived, the frequency-integrated PL (broad band) and the frequency-resolved PL (narrow band) can be readily obtained. At this regard, we observe that the Heisenberg exciton operator $\hat{B}_\alpha (t)$ describes a local oscillating dipole within the quantum well. Therefore, it is the source of an emitted electromagnetic wave that is related to the sample polarization:

$$P_k (t) = \langle \hat{E}_k^\dagger (t) \rangle = \sum_\alpha M_\alpha^* (k) \langle \hat{B}_\alpha^\dagger (t) \rangle \quad (8.45)$$
and the time-resolved PL intensity can be written as

\[ I_k(t) = \left\langle \hat{E}_k^\dagger(t) \hat{E}_k(t) \right\rangle = \sum_{\alpha\beta} M_{\alpha}^* (k) M_{\beta} (k) \left\langle \hat{B}_\alpha^\dagger(t) \hat{B}_\beta(t) \right\rangle. \]  

(8.46)

Taking into account diagonal terms only, the most relevant ones in PL spectra, the frequency-integrated PL intensity can be promptly written as

\[ I_k(t) = \left\langle \hat{E}_k^\dagger(t) \hat{E}_k(t) \right\rangle = \sum_\alpha |M_\alpha(k)|^2 \left\langle \hat{B}_\alpha^\dagger(t) \hat{B}_\alpha(t) \right\rangle = \sum_\alpha |M_\alpha(k)|^2 N_{\alpha\alpha}(t) \]  

(8.47)

and the frequency-resolved PL intensity has the expression

\[ I(\omega_d) = \sum_\alpha |M_\alpha(k)|^2 N_{\alpha\alpha}(t) \frac{1}{i(\omega_\alpha - \omega_d - i\Gamma_\alpha/2)}. \]  

(8.48)

If we consider a spatially confined detection, we must take into account the overlap of the exciton wave functions with the signal mode \( E_C(r) \) delivered by the tip in order to obtain the signal that can be detected by a general near-field setup (collection mode). Using strictly similar expressions to (8.15 - 8.17), we can write the spatially-resolved, frequency-integrated PL intensity as

\[ I_{Ck}(t) = \sum_\alpha |E_{C\alpha}|^2 N_{\alpha\alpha}(t) \]  

(8.49)

and the spatially-resolved, frequency-resolved PL intensity as

\[ I_C(\omega_d) = \sum_\alpha |E_{C\alpha}|^2 N_{\alpha\alpha}(t) \frac{1}{i(\omega_\alpha - \omega_d - i\Gamma_\alpha/2)}, \]  

(8.50)

both depending upon the collecting tip position.  

As for the case of a near-field setup we shall need to consider the diagonal occupation equation with the proper input and output fields, it becomes

\[ \partial_t N_\alpha = G_\alpha(\omega_{in}) + \sum_\beta W^{(1)}_{\alpha\beta} N_\beta - \Gamma_\alpha N_\alpha, \]  

(8.51)
where $N_\alpha$ are the diagonal terms of the exciton density matrix. In this equation the generation term depends on the spatial overlap between the illuminating beam and the exciton wave functions corresponding to exciton levels resonant with the input light:

$$G_\alpha = r_0 |o^{in}_\alpha|^2 \mathcal{L}_\alpha(\omega_{in}),$$

(8.52)

with $\pi \mathcal{L}_\alpha(\omega) = (\Gamma_\alpha/2)/[(\omega - \omega_\alpha)^2 + (\Gamma_\alpha^2/4)]$ and $o^{in}_\alpha = \int d^2 \mathbf{R} \tilde{E}_{in}(\mathbf{R}) \psi_\alpha(\mathbf{R})$, where $\tilde{E}_{in}(\mathbf{R}) = \int E_{in}(\mathbf{r}) f(z) dz$, $\Gamma$ is the dephasing rate of the exciton due to radiative decay emission and phonon scattering.

Once the exciton densities have been derived, the frequency integrated PL can be readily obtained. It results:

$$I(\bar{\mathbf{R}}_{out}) = r_0 \sum_\alpha |o^{out}_\alpha(\bar{\mathbf{R}}_{out})|^2 N_\alpha,$$

(8.53)

where $o^{out}_\alpha$, analogously to $o^{in}_\alpha$, contains the overlap of the exciton wavefunctions with the signal mode $\tilde{E}_{out}(\mathbf{R})$ delivered by the tip centered at $\bar{\mathbf{R}}_{out}$ (collection mode). According to the quantum regression theorem, $\langle \hat{S}^-(0) \hat{S}^+(\tau) \rangle$ has the same dynamics of $\langle \hat{S}^+(\tau) \rangle$ (proportional to the exciton operator), but with $\langle \hat{S}^-(0) \hat{S}^+(0) \rangle$ as initial condition. Following this procedure we obtain

$$I_{PL}(\bar{\mathbf{R}}_{out}, \omega_{out}) = r_0 \sum_\alpha |o^{out}_\alpha(\bar{\mathbf{R}}_{out})|^2 \mathcal{L}_\alpha(\omega_{out}) N_\alpha.$$  

(8.54)

We apply the above developed theoretical scheme to calculate the individual occupations of exciton states confined in the dot after a continuous-wave far-field optical excitation resonant with the energy of the 1s QW exciton (the dot barrier). The obtained occupations are then used to study the spatially and spectrally resolved (collection mode) light emission from the dot. The effective potential felt by 1s excitons used in our simulations is represented by a sample of $(240 \times 240)$ nm with a prototypical interface-fluctuation confinement of rectangular shape with dimensions $(60 \times 90)$ nm, and monolayer fluctuations giving rise to a 6 meV effective confinement potential.
8.1.4 Simulated experiments

We apply the above developed theoretical scheme to calculate the individual occupations of exciton states confined in the dot after a continuous-wave far-field optical excitation resonant with the energy of the $1s$ QW exciton (the dot barrier). The obtained occupations are then used to study the spatially and spectrally resolved (collection mode) light emission from the dot. The effective potential felt by $1s$ excitons used in our simulations is represented by a sample of $(240 \times 240)$ nm with a prototypical interface-fluctuation confinement of rectangular shape with dimensions $(60 \times 90)$ nm, and monolayer fluctuations giving rise to a 6 meV effective confinement potential. The upper panel of Fig. 8.1 displays the near-field PL signal as a function of photon energy and beam position obtained after uniform illumination of the sample at the energy of the QW $1s$ exciton. $T=2$ K, FWHM $=40$ nm. (b) Total absorption under local illumination, along the same line, spatial resolution and temperature as in (a).

Figure 8.1. (color online). (a) Near-field PL signal (collection-mode) as a function of photon energy and beam position (line scan shown in the inset) obtained under uniform illumination of the sample at the energy of the QW $1s$ exciton. $T=2$ K, FWHM $=40$ nm. (b) Total absorption under local illumination, along the same line, spatial resolution and temperature as in (a).
of the sample at energy $\omega_I = 0$ meV (the zero is fixed at the energy of the 1s QW exciton in the absence of interface fluctuations). The specific line scan is indicated in the inset. A Gaussian profile with FWHM = 40 nm of the electromagnetic-mode supported by the collecting tip has been assumed. The line scan clearly evidences the first excited state of the dot which is dark under far-field collection. We observe that its spectral line is more intense and narrow (owing to the absence of radiative decay) than that of the ground state. The calculated PL spectra shows that dark-states can be observed by high-resolution SNOM in the usual collection mode configuration after nonresonant far-field excitation, without the need of nonlinear optical interactions. Analogous results are expected for locally collected electroluminescence. For comparison we plotted in the lower panel the total absorption under local illumination (FWHM = 40 nm). As near-field PL is proportional to this quantity times the level occupations, the comparison provides interesting information about the steady-state exciton populations. The two panels clearly indicate that the level occupation of the dark-state is significantly larger than that of the ground state, at the opposite to what predicted by the Boltzmann distribution. The observed inverted occupations origin from symmetry-suppression of radiative decay of the dark-state and the quite small nonradiative scattering at low temperature ($T = 2K$) for states well confined in the dot. This behaviour is better evidenced in panels 8.2(a) and 8.2(b). The spectra have been obtained centering the tip at one of the two emission maxima of the first excited state (LS) and at the emission maximum of the ground state (C). In order to better specify the origin of the inverted occupations, we calculated the PL spectra as in Fig. 8.2(a) except that we artificially set the radiative decay rates of all the levels two orders of magnitude lower than the lowest nonradiative decay rate. Panel 8.2(c) displays the results showing that, as expected in this case, equilibrium is recovered and the near-field emission from the dot lowest energy level dominates even when the tip is centered on the maximum
Figure 8.2. (color online). (a) PL spectra obtained in collection mode ($FWHM = 40$ nm and $T = 4$ K). The spectra have been obtained centering the tip at one of the two emission maxima of the first excited state (LS) and at the emission maximum of the ground state (C); (b) Total absorption under local illumination obtained with spatial resolution, at temperature and centering the snom tip as in Fig. 8.2(a); (c) PL spectra as in Fig. 8.2(a) where the radiative decay rates of all the levels are set two orders of magnitude lower than the lowest nonradiative decay rate. (d) Level occupations (normalized at the maximum) calculated for two different temperatures.

of the dark-state emission. Fig. 8.2(d) shows the calculated level occupations at $T = 2K$ and $T = 30K$. At $T = 2K$ all the occupations with the striking exception of the second energy level decrease monotonically with energy. This level displaying an occupation density which is more then a factor two that of the lowest energy level. At $T = 30K$ the monotonous behaviour is recovered for all the energy levels. We notice that the sum of the occupations obtained at $T = 30K$ is significantly larger then that at $T = 2K$. At low temperature a large fraction of the resonantly generated QW excitons (with a quite large radiative decay rate) recombine by radiative emission, while at higher temperature phonon scattering lowers this effect, increasing the carrier density captured by the dot. Fig. 8.3 shows the near-field
(FWHM = 40 nm) PL spectral line-scans obtained at three different temperatures. Increasing the temperature all states are able to better thermalize, frequently emitting and absorbing phonons before radiative emission. The influence of spatial resolution on the PL spectral line-scans is shown in Fig. 8.4. This figure displays the line-scans calculated at $T = 2K$ for three different spatial resolutions (indicated in the figure). Lowering spatial resolution, the signal from the dark state tend to disappear owing to cancellation effects in the matrix element $\sigma^\text{out}_\alpha$ governing the light-matter coupling. Finally, we test the influence of random interface fluctuations with a correlation length of the order of the exciton Bohr-radius. Fig. 8.5(a) displays the
specific realization of the effective disordered potential used for the calculation of the PL image. This potential is obtained adding to the dot potential used for all the previous calculations a contribution modeled as a zero mean, Gauss distributed and spatially correlated process with a correlation length $\sigma = 8$ nm and a width of the energy distribution $v_0 = 0.6$ meV (about the 10% of the dot potential-well). Fig. 8.5(b) displays the energy-integrated PL image ($FWHM = 40$ nm, $T = 2$K). It evidences the efficiency of the dot capture and near-field emission. Fig. 8.5(c) shows that the presence of the symmetry-breaking disordered potential slightly increases the linewidth of the second energy level, determining a slight equilibration of the
Figure 8.5. (color online). (a) Specific realization of the effective disordered potential used for the calculation of the PL images and spectra show. (b) PL energy-integrated image obtained after uniform illumination of the sample at the barrier energy and collecting locally ($FWHM = 40$ nm, $T = 4$ K). (c) PL spectra calculated centering the tip in the center of the dot and in the position indicated by a circle in the first panel ($FWHM = 40$ nm, $T = 4$ K). (d) Far-field PL spectrum ($FWHM = 40$ nm, $T = 4$ K).

emission lines. Fig. 8.5(d) displays the far-field PL spectrum. A very small peak (not present in the absence of disorder) appears at the energy of the second level, which thus remains mainly dark, demonstrating the robustness of dark-states with respect to this kind of disorder. Results obtained for other specific realizations of the random potential (here not shown), don’t display qualitative differences.
Part IV

Quantum optics with cavity photons
Chapter 9

Decoherence-Free Emergence of Macroscopic Local Realism

Entanglement is one of the most profound features of quantum mechanics. It plays an essential role in all branches of quantum information theory [112]. Bell theorem [113], which is derived from Einstein-Podolsky-Rosen’s (EPR’s) notion of local realism [114], quantifies how measurements on entangled quantum mechanical systems may invalidate local classical models of reality. While all bipartite pure entangled states violate some Bell inequality [115], the relationship between entanglement and non-locality for mixed quantum states is not well understood yet [116, 117]. Moreover recent proposals [1] and realizations [2, 3, 4, 5, 6] of many-particle entangled quantum states require a better understanding of the domain of validity of quantum behaviour. In the literature two main research lines concerning entanglement can be easily found, one more focused on the group-theoretic properties of the Hilbert space under investigation and the other more interested in which feasible measurement may quantify nonclassical and/or entanglement properties. We shall move our considerations within this second paradigm. A relevant point is whether the conflict between classical elements of reality and quantum mechanics may persist.
at a macroscopic level [33, 34]. Indeed continuous-variable entanglement of intense light sources has been recently demonstrated in [2, 4] and polarization entanglement of macroscopic beams in [5]. It has been recently shown that a source of strongly entangled states with photon numbers up to a million seems achievable in [1]. In these works entanglement has been tested and quantified by means of specific separability criteria which are inequalities among expectation values of experimentally measurable quantities, violated by entangled quantum states [118]. The behaviour of entanglement towards a macroscopic situation (even close to classical every-day life phenomena) and its robustness versus noise and decoherence are not well understood and the quantum-to-classical transition is usually associated with decoherence [34], i.e. the practically unavoidable spreading of information from the system to the environment. In this Chapter we shall address this crucial problem focusing on a particular very promising source of macroscopic entanglement: parametric down-conversion of photons inside an optical cavity.

![Figure 9.1. Polariton energy versus the in-plane wave vector. The parametric polariton-polariton process generating signal and idler polariton pairs is also depicted.](image-url)
It is worth mentioning there is a somewhat close analogy between parametric
down conversion of photons by BBO crystal in an optical cavity and spontaneous
parametric emission of polaritons in a SMC which, thanks to polariton-polariton
parametric scattering, can create pairs of quantum-correlated signal/idler polariton
pairs, see Fig. 9.1. The great advantage of these structures is their ability in
producing, easily, polarized-entangled many-body quantum state. Figure 2 of Ref.
[12] shows the integrated outgoing photoluminescence versus the pump density, it
is clear that after a quadratic region for very low pump density a threshold occurs,
beyond the behaviour becomes exponential witnessing a parametric phenomenon is
taking over inside the SMC. There are some works [21, 119] which are exploiting
this analogy and this thesis also is mostly dedicated to build a bridge between those
quantum optics experiments of cavity quantum electrodynamics with photons in an
optical cavity and ultrafast spectroscopy in SMCs.

Following Ref. [35], on the one hand we shall quantify the detrimental influence
of such environment channels and show how self-stimulation may suppress them
efficiently. On the other hand we shall tackle the problem of the macroscopic limit
and of the emergence of classical elements of reality within a quantum framework.
We shall illustrate a counter-example where the emergence of macroscopic local
realism (MLR) may be seen as an intrinsic feature of quantum systems, endogenous
in the quantum theory itself (even in the presence of strong entanglement that is
the quintessential of non-classicality). In such a case there is no need at all to rely
on environment ingredients (like noise and decoherence). Our results, of course,
do not imply that macroscopic entangled systems cannot display violations of local
realism [36, 37], but that there is a large class of quantum correlation measurements
that cannot be used to show them. This is a great debated topic because, actually,
inequalities made up with these correlations are the most used mean for testing
and even prove the non-classical, i.e. quantum, features of famous experiments
[38, 39]. In addition, also for continuous-variable teleportation some authors [40] are wondering whether a truly quantum description is needed or an explanation in terms of classical phase-space correlation suffices. We shall firstly deal with a specific situation which is the Heisenberg steady-state Eq. (9.2), this way we are able to focus on the most important physical ingredients in a very neat way. It is worth noting that this does not restrict our conclusions at all, indeed we prove that in the more realistic t-dependent Langevin approach of Eq. (9.8) all our results continue to hold (see fig. 9.3). The results presented here indicate that MLR may result from the inability of the observer, practically unavoidable for macroscopic systems, to catch the quantized structure of the system. In order to investigate the relationship among entanglement, quantum nonlocality, and the macroscopic limit, we adopt a weaker nonlocality concept [120] based on an adherence on the physical system. In particular we do not ask quantum correlations to exceed bounds that cannot be violated by classical correlations, but we limit us to compare the quantum findings with the corresponding classical stochastic calculations for the specific physical system under investigation. However, a classical model endowed with stochastic noise represents a realization of a local stochastic hidden variable theory and the customary lines of thoughts are recovered.

We consider polarization entangled light from parametric down-conversion driven by an intense pump field inside a cavity. The multiphoton states produced are close approximations to singlet states of two very large spins [1]. The interaction Hamiltonian describing the process is given by

\[ \hat{H} = i\hbar \Omega (\hat{a}_h^\dagger \hat{b}_v - \hat{a}_v^\dagger \hat{b}_h) + H.c., \]  

(9.1)

where \( a \) and \( b \) refer to the two conjugate directions the frequency-degenerate photon pairs are emitted along. \( h \) and \( v \) denote horizontal and vertical polarization and \( \hbar \Omega \) is a coupling constant whose magnitude depends on the nonlinear coefficient of the crystal and on the intensity of the pump pulse. In the absence of losses, within
the Heisenberg picture, the interaction Hamiltonian in (9.1) dictates the following steady-state solution for photon operators:

\[
\hat{a}_{h,v} = \hat{a}_{h,v}(0) \cosh(r) \pm \hat{b}_{v,h}(0) \sinh(r) \\
\hat{b}_{v,h} = \hat{b}_{v,h}(0) \cosh(r) \pm \hat{a}_{h,v}(0) \sinh(r),
\]

where the interaction parameter \( r \) is \( \Omega \tau \) being \( \tau = L/v \) the interaction time interval, i.e. the time spent by the photons with velocity \( v \) inside a crystal of length \( L \).

In the absence of losses and considering the photon vacuum as initial state, the Hamiltonian (9.1) produces a multiphoton quantum state \( |\psi\rangle \) that is the polarization equivalent of a spin singlet state, where the spin components correspond to the Stokes polarization parameters, \( \hat{J}_Z^A = (\hat{a}_h^\dagger \hat{a}_h - \hat{a}_v^\dagger \hat{a}_v)/2, \) \( \hat{J}_Z^B = (\hat{a}_+^\dagger \hat{a}_+ - \hat{a}_-^\dagger \hat{a}_-)/2, \) and \( \hat{J}_y = (\hat{a}_l^\dagger \hat{a}_l - \hat{a}_r^\dagger \hat{a}_r)/2, \) where \( \hat{a}_{+,-} = (\hat{a}_h \pm \hat{a}_v)/\sqrt{2} \) correspond to linearly polarized light at \( \pm 45^\circ \), and \( \hat{a}_{l,r} = (\hat{a}_h \pm i\hat{a}_v)/\sqrt{2} \) to left- and right-ended circularly polarized light. The label \( A \) refers to the \( a \) modes. analogous expressions can be obtained for \( \hat{J}_B \) in terms of the \( b \) modes. It has been shown [1] that the state \( |\psi\rangle \) is a singlet state of the total angular momentum operator \( \hat{J} = \hat{J}_A + \hat{J}_B \). As a consequence \( \langle \psi | \hat{J}^2 | \psi \rangle = 0 \). Losses, fluctuations and imperfections may lead to nonzero values for the total angular momentum, corresponding to nonperfect correlations between the Stokes parameters in the \( a \) and \( b \) beams. Within this picture it is straightforward to include the presence of noise in the system assuming that, before the pump is switched on, the system is in an incoherent thermal-like state described by a diagonal density matrix. Dealing with such systems the first analysis one may perform is an intensity measurement:

\[
n_{ah(v)}(r) \equiv \langle \hat{a}_{h,v}^\dagger \hat{a}_{h,v} \rangle = \sinh^2 r + n_o(1 + 2 \sinh^2 r),
\]

where \( n_o = \langle \hat{a}_{h,v}(0) \hat{a}_{h,v}(0) \rangle = \langle \hat{b}_{h,v}(0) \hat{b}_{h,v}(0) \rangle \) is the noise present in the system. There are two different contributions: the first term arises from vacuum fluctuations and describes true (eventually self-stimulated) spontaneous emission, vanishing in
the absence of parametric interaction; the latter describes a classical-like amplification of the input thermal noise \( n_0 \). It is worth noting that the solution of the corresponding classical model of an optical parametric amplifier has the same structure of Eq. (9.2) with \( a \) and \( b \) being of course replaced by classical amplitudes [121]:

\[
n^C(r) \equiv \langle a^{\ast}_{h,v} a_{h,v} \rangle = n_0^C(1 + 2 \sinh^2 r),
\]

(9.4)

where \( \langle \rangle \) denotes statistical average and \( n_0^C \) is, as before, statistical noise. For small \( r \) values and for negligible \( n_0 \) (\( n_0 << r^2 \)), quantum and classical descriptions lead to distinct behaviour in \( r \), being \( n(r) \simeq r^2 \) and \( n^C(r) \simeq n^C(0) \). It can be related to the fact that vacuum fluctuations (in contrast with classical ones) do not produce work and hence, while they can stimulate pump scattering, they cannot be directly evidenced by photodetection. In contrast when \( r \) increases, it is no more possible to identify quantum behaviour by means of simple intensity measurements. In particular for \( r \geq 2 \) a classical model with \( n_0^C = n_0 + 1/2 \) (in order to properly include vacuum fluctuations), is able to give an intensity description that cannot be distinguished from the quantum one. It is worth noting that this behaviour can also be found in intriguing second-order interference effects [120, 122] and it agrees with the old idea that many quanta in a system give rise to a classical-like behaviour. Other relevant second order correlations are given by the following anomalous correlators:

\[
A_{hv(vh)} = \langle \hat{a}_{h(v)} \hat{b}_{v(h)} \rangle = (n_0 + 1/2) \sinh(2r)
\]

\[
A_{h^v(v^h)}^C = \langle a_{h(v)} b_{v(h)} \rangle = n_0^C \sinh(2r),
\]

(9.5)

which quantify the pair correlation induced by the parametric process. Equation (9.5) shows that replacing again \( n_0^C = n_0 + 1/2 \), the classical description coincides with the quantum one.

If the above hold for second-order correlations, now we want to focus our attention on what we can say about entanglement measurements on this system. In
order to test the degree of entanglement, a simple inseparability criterion has been
derived [1]: if \( \frac{\langle \hat{J}^2 \rangle}{\langle \hat{N} \rangle} \) (where \( \hat{N} = \hat{N}_A + \hat{N}_B \) is the total photon number) is
smaller than \( \frac{1}{2} \), then the state is entangled. We now consider the system at \( r \leq 0 \)
(before switching on the pump) to be in thermal equilibrium, i.e. in a completely
incoherent (mixed) state described by a diagonal density matrix. The only input for
the system are thermal noise (if \( T \neq 0 \)) and vacuum fluctuations. From Eq. (9.2)
we obtain:

\[
\frac{\langle \hat{J}^2 \rangle}{\langle \hat{N} \rangle} = \frac{3n_0(n_0 + 1)}{4n_0 + (1 + 5n_0) \sinh^2 r}.
\] (9.6)

Figure 9.2. (color online). Separability criterion for different noise versus the in-
teraction parameter \( r \). At zero temperature \( n_0 = 0 \) and the system is maximally
entangled independently of the magnitude of \( r \). For \( r \rightarrow \infty \) according to the crite-
ron macroscopic entanglement may in principle be achieved even in the presence of
strong fluctuations, provided that self-stimulation of the emitted pairs takes place.

Figure 9.2 shows the behaviour of eq. 9.6 different noise values versus the interac-
tion parameter \( r \). At zero temperature \( n_0 = 0 \) and the system is maximally entangled
(\( \langle \hat{J}^2 \rangle = 0 \) independently of the magnitude of \( r \). As Eq. (9.6) shows, even when
thermal noise largely exceeds vacuum fluctuations (\( n_0 >> 1 \)), \( \langle \hat{J}^2 \rangle / \langle \hat{N} \rangle \) goes
below $1/2$ provided $r$ being large enough. Moreover $\langle \hat{J}^2 \rangle / \langle N \rangle \rightarrow 0$ for $r \rightarrow \infty$. Thus macroscopic entanglement may in principle be achieved even in the presence of strong fluctuations, provided that self-stimulation of the emitted pairs takes place. In particular the system becomes entangled when $\sinh^2 r > 2n_0(3n_0 + 1)/(5n_0 + 1)$. Nevertheless, according to the criterion, in order to beat the detrimental effect of strong fluctuations on entanglement we need to rely on self-stimulation. It is known that entanglement as well as violations of Bell inequalities have limited resistance to noise. Here a small amount of noise is enough to completely destroy entanglement, e.g. for $r = 10^{-3}$, $n_0 = 2 \times 10^{-6}$ is sufficient to wash out entanglement according to Eq. (9.6); nevertheless switching on self-stimulation (by increasing $r$) we shall restore it. In order to get a deeper insight we seek some additional information wondering if, from the criterion viewpoint (this time), we can distinguish between classical and quantum findings. To this end we put the two descriptions (classical and quantum) on equal footing and compute the entanglement criterion evaluating their differences and similarities. A classical calculation, computed according to the above described prescriptions, gives $\langle \hat{J}^2 \rangle / \langle \hat{N} \rangle = 3n_0^c/(4 + 5 \sinh^2 r)$. Of course classical optics does not require a minimum amount of fluctuations, thus within a classical model it is possible to obtain $\langle \hat{J}^2 \rangle / \langle \hat{N} \rangle < 0.5$. In the low excitation regime ($r << 1$) and $n_0$ lower than $r^2$ classical and quantum calculations of $\langle \hat{J}^2 \rangle / \langle \hat{N} \rangle$ display very different variations with $r$ as it happens for simpler intensity cases. As $r$ increases they tend to coincide. This means that experiments eventually demonstrating macroscopic entanglement for this system can be accounted for in terms of purely classical correlations, with no need for a quantum-mechanical explanation. Analogous conclusions can be reached for different experimentally tested criteria [2, 4, 5, 6]. This does not mean at all that the entanglement criterion is wrong or contradictory. In contrast to Bell’s inequalities, these kind of criteria are derived exploiting the fact that involved operators do not commute (hence they do not hold
for classical descriptions). Also these results do not imply that macroscopic entangled systems cannot display quantum nonlocality effects. Indeed it has been shown in [36] that quantum states of a nondegenerate optical parametric amplifier display violations of the Bell inequality due to Clauser, Horne, Shimony, and Holt in the macroscopic limit \((r \to \infty)\). However the above analysis suggests that there is a large class of quantum correlation functions that cannot differ from the corresponding classical ones in the macroscopic limit. Indeed we can define the following set of correlation functions

\[
\langle \hat{B}_\alpha^{(n)} \rangle, \text{ where } \hat{B}_m = (\hat{b}_v)^{n-m}(\hat{a}_v)^{m-l}(\hat{a}_h)^{l-k},
\]

is a generic \(n\)-particle destruction operator. These correlation functions (and also their classical counterparts \(\langle B^{(n)} \rangle\)) are different from zero only if \(n = n'\) and \(\alpha \equiv (k,l,m) = \alpha'\). Since we are dealing with a Gaussian system [123], such correlators (quantum and classical) can be decomposed in a sum of products of second order correlation functions only \((n(r)\) and \(A(r)\). Since \(A(r) = A^C(r)\) (for \(n^C_0 = n_0 + 1/2\)), and \(n(r)/n^C(r) \to 1\) for \(r \to \infty\), we obtain that

\[
\lim_{r \to \infty} \frac{\langle \hat{B}_\alpha^{(n)} \hat{B}_\alpha^{(n')} \rangle}{\langle B^{(n)} B^{(n')} \rangle} = 1. \tag{9.7}
\]

This result implies that it is not possible to observe violations of macroscopic local realism (e.g. violations of Bell inequalities, including those which are not yet known) by measurements of any finite set of expectation values that can be expanded as a finite sum of these correlation functions. It is easy to verify via explicit calculations that convergence of limit (9.7) is very fast even for large values of \(n\). Bell inequalities can be schematically expressed as \(\mathcal{F}(\langle B^{(n)} B^{(n')} \rangle (r)) \leq \mathcal{L}(n)\), where \(\mathcal{L}(n)\) is a bound imposed by local realism that cannot be violated by classical correlations, and \(\mathcal{F}\) is a generic continuous function of \(\langle B^{(n)} B^{(n')} \rangle\) depending also on the different settings chosen by the observers. From Eq. (9.7): \(\mathcal{F}(\langle \hat{B}_\alpha^{(n)} \hat{B}_\alpha^{(n')} \rangle (r)) \to \mathcal{F}(\langle B^{(n)} B^{(n')} \rangle (r))\) when \(r \to \infty\), thus any bound \(\mathcal{L}\) cannot be violated (in the limit). One example of these wide class of Bell inequalities can be found in [124]. We stress that Eq. (9.7) has been obtained simply by exploiting Gaussian factorization.
and comparing the macro-limit for classical and quantum second-order correlations; it is thus clear that it can be easily generalized to a wide variety of Gaussian systems with larger degrees of freedom, e.g. to multipartite situations obtained by inserting in the setup a number of beamsplitters [125].

So far we have considered a situation where the system is initially in thermal equilibrium, but Eq. (9.2) has been obtained under the hypothesis that the system has no losses, hence it is assumed that for the steady-state calculations at any value of the interaction parameter \( r \) the system is disconnected from the environment. However in real systems amplification, losses, and noise disturbances actually take place simultaneously. Thus (steady-state) Heisenberg equations have to be replaced by (t-dependent) Langevin equations with noise sources. In the symmetric case (equal losses for all the four modes), we obtain,

\[
\hat{a}_h(t) = \hat{a}_h(0)e^{-\lambda t} \cosh \Delta(t,0) + \hat{b}_v(0)e^{-\lambda t} \sinh \Delta(t,0) + \int_0^t e^{-\lambda(t-t')} \cosh \Delta(t,t') \hat{f}_{ah}(t') dt' \\
+ \int_0^t e^{-\lambda(t-t')} \sinh \Delta(t,t') \hat{f}_{bv}(t') dt'
\]

(9.8)

where \( \Delta(t,t') = \int_0^t \kappa(t'') dt'' = \frac{n_0}{\Lambda}(e^{-\Lambda t'} - e^{-\Lambda t}) \); \( \hat{f}_\alpha(t) \) are Bose quantum noise operators associated with the losses [126] (\( \alpha \) denotes the specific mode e.g. \( \alpha \equiv (a,h) \)).

In the following we will assume \( \langle \hat{f}_\alpha(t)\hat{f}_{\alpha'}(t') \rangle = 2\lambda n_0 \delta_{\alpha,\alpha'} \delta(t-t') \). Analogous expressions can be obtained for the other three modes. Fig. 9.3 displays the quantum and the classical calculation of \( \langle \hat{J}_2^2 \rangle / \langle N \rangle \) for \( \lambda = \Lambda = 0.1, \kappa_0 = 1 \). For the quantum (continuous line) and the classical (dashed line) calculations we adopted \( n_0 = 0.3 \) and \( n_0^{(d)} = 0.8 \) respectively. The figure clearly shows that for \( r > 3 \) classical and quantum results cannot be distinguished. Thus i) also in this more realistic case self-stimulation can suppress the detrimental effect of noise, ii) coincidence between classical and quantum results can be obtained without requiring \( n >> 1 \) if we choose \( n_0^{(d)} = n_0 + 1/2 \); iii) decoherence due to losses and noise seems to affect
equally quantum entanglement and classical correlations, hence it cannot be invoked in the present case for the emergence of a classical behaviour. In order to interpret our results, we distinguish between two situations: when \( r << 1 \), the probability to deal with states with more than two-photons is negligible, so measurements of \( \langle \hat{J}^2 \rangle \) and of \( \langle \hat{N} \rangle \) can probe the system at a microscopic level, but with a lot of particles (when \( r \) increases) they both become macroscopic observables unable to probe the system fluctuations with precision at a few quanta level. In this case the information recovered by observations is a coarse grained quantity missing the underlying quantum structure. This lack of microscopic information seems able to introduce elements of local realism even in the presence of strong entanglement and in the absence of decoherence. Of course the lack of information here described should not be confused with a lack of precision of measurements, here assumed with unlimited precision. As shown in Ref. [127], the partition of a quantum system into subsystems...
and hence the entanglement structure, is dictated by the set of operationally accessible interactions and measurements. A given set can hide a multipartite structure. Our results suggest that, analogously, the set of operationally accessible measurements and their ability to catch the quantized structure of the system determine the quantum or classical nature of the observed correlations. As pointed out above, our findings (including Eq. (9.7)) do not imply that macroscopic entangled systems cannot display quantum nonlocality effects. As already mentioned, it has been shown that these kind of systems do violate CHSH Bell inequality in the macroscopic limit ($r \to \infty$) [36]. However in that case the Bell operator is constructed by means of operators with single quantum sensitivity independently of the number of particles in the system in contrast to operators $\hat{B}_\alpha^{(n)}\hat{B}_\alpha^{(n)}$. Of course these operators cannot be expanded in a finite sum of operators $\hat{B}_\alpha^{(n)}\hat{B}_\alpha^{(n)}$. Analogous conclusions can be drawn for the violations of macroscopic local realism shown in [33].

The emergence of macroscopic local realism in the presence of strong entanglement, shown here, provides insight into the boundary between classical and quantum worlds. These results, with the care that they have been obtained for a Gaussian system, suggest that, despite the feasible realization of systems with a huge amount of entangled particles, the lack of information gathered by coarse-grained observations may lead to the introduction of elements of local realism even in the presence of strong entanglement and in the absence of decoherence. In particular, Eq. (9.7) shows that by using apparata able to measure finite-order correlations only, it is not possible to detect violations of local realism for macroscopic Gaussian states. This is a great debated topic because, actually, inequalities made up with these correlations are the most used mean for testing and even prove the non-classical, i.e. quantum, features of famous experiments [38, 39]. In addition, also for continuous-variable teleportation some authors [40] are wondering whether a truly quantum description is needed or an explanation in terms of classical phase-space correlation suffices.
Further investigations are needed to understand if our results can be extended to more general quantum systems.
Part V

Conclusions and outlook
Chapter 10

Conclusions and outlook

The main aim of this thesis had been to try to contribute to develop a theoretical framework able to describe optical response in semiconductor heterostructures both in the weak- and strong-coupling regimes. All the similarities between quantum optics in atom-cavity systems and polaritons in semiconductor microcavities lead us to ask ourselves if both these disciplines would benefit from a fruitful intertwined relationship, indeed we are confident that each of them would acquire the techniques, the knowledge, the interests and the open questions of the other one. One very hot topic in many research areas nowadays is entanglement as a key tool in quantum information and communication technology [10]. Indeed parametric down-conversion (PDC) is the most frequently used method to generate highly entangled pairs of photons for quantum-optics applications, such as quantum cryptography and quantum teleportation. Moreover, the rapid development of these disciplines asks for monolithic, compact sources of nonclassical photon states enabling efficient coupling into optical fibres and possibly electrical injection. Semiconductor-based sources of entangled photons would therefore be advantageous for practical quantum technologies.
In part IV we addressed the relevant point whether the conflict between classical elements of reality and quantum mechanics may persist at a macroscopic level [33, 34] and if quantum features themselves — such as entanglement — can be created or founded even among macroscopic objects too. Focusing on a very promising source of macroscopic entanglement, PDC, we had tackled the problem of the macroscopic limit and of the emergence of classical elements of reality within a quantum framework providing a counter-example where the emergence of macroscopic local realism (MLR) may be seen as an intrinsic feature of quantum systems, endogenous in the quantum theory itself even in the presence of strong entanglement worldwide accounted for as a firm mark of non-classicality, with no need at all to rely on decoherence for the quantum system to mock classical reality as well. In order to be as clear as possible it seems worth to underline that our results do not imply that macroscopic entangled systems cannot display violations of local realism [36, 37], but that there is a large class of quantum correlation measurements that cannot be used to show them. With the care that they have been obtained for a Gaussian system, our findings suggest that, despite the feasible realization of systems with a huge amount of entangled particles, the lack of information gathered by coarse-grained observations may lead to the introduction of elements of local realism even in the presence of strong entanglement and in the absence of decoherence.

The strong light-matter interaction in semiconductor microcavity systems gives rise to cavity polaritons which are hybrid quasiparticles consisting of a superposition of cavity photons and quantum well excitons [11]. Demonstrations of parametric amplification and parametric emission in SMCs [12, 13, 14], together with the possibility of ultrafast optical manipulation and ease of integration of these microdevices, have increased the interest on the possible realization of nonclassical cavity-polariton states with a very high degree of entanglement. In part II after developing a DCTS
theoretical framework for interacting polaritons, we have presented a general theoretical approach for the realistic investigation of polariton quantum correlations in the presence of coherent and incoherent interaction processes. The theory joins the dynamics controlled truncation scheme and the non-equilibrium quantum Langevin approach to open systems. It provides an easy recipe to calculate multi-time correlation functions which are key quantities in quantum optics, but, as shown, even for single-time quantities it provides an easy and advantageous decoupling of incoherent dynamics from parametric processes. As a first application of the proposed theoretical scheme, we have analyzed the build-up of polariton parametric emission in semiconductor microcavities including the influence of noise originating from phonon induced scattering. Our numerical results clearly show the importance of a proper microscopic analysis able to account for parametric emission and pump-induced PL on an equal footing in order to make quantitative comparison and propose future experiments, seeking and limiting all the unwanted detrimental contributions. Specifically, we have shown that already at moderate pump excitation intensities there are clear evidences that we may device future practical experiments exploiting existing situations where the detrimental pump-induced PL contribution is very low meanwhile we face a good amount of polaritons per mode. It represents an exciting and promising possibility for future coincidence experiments even in photon-counting regimes, vital for investigating (quantum) coherence properties of the emitted light. However, it has been shown theoretically [79] that it is quite difficult to populate the polaritons in the strong coupling region by means of phonon-scattering due to a bottleneck effect and, actually, this effect is experimentally observed only very partially and under particular circumstances [80]. This is mainly due to other more effective scattering mechanisms [81] usually present in SMCs such as the interaction with free electrons. Our results do not display arbitrary units and our scheme has the great ability to include quite naturally these other enriching contributions pushing
the numerical results even closer to the experimental evidences. As a consequence, it would provide in a near future a powerful microscopic tool able to analyze existing experiments, proposing meanwhile innovative future investigations to compare the results with. In particular, for very common experimental conditions [128] polariton parametric scattering is supposed to generate pairs of spin-entangled polaritons. We are currently developing our present theoretical model in order to include the spin in the model and the proper description of coincidence detections which are the key quantities in experiment aiming at quantify polariton (spin) entanglement properties in these systems. Moreover parametric processes in SMC are expected to give a massive and easy production of entangled multiparticle polariton states. Multiparticle entanglement is of great interest in quantum optics and quantum information theory and a lot of effort is being made for a better understanding of its subtle properties. Exploiting and developing our theoretical scheme would allow us in a near future to devise and propose feasible set-ups where such a polariton entanglement lasing could be theoretically predicted and experimentally demonstrated.

Optical spectroscopy of ultrafast dynamics has become in recent years a strategic field both in research and from a technological point of view. The achievement of very high spatial and temporal resolutions in optical spectroscopies of molecules and solids is among the important experimental advancements of recent years. With near-field scanning optical microscopy (SNOM), the spatial resolution is reduced below the diffraction limit and approaches the scale of quantum confinement [28]. The matrix elements governing the light-matter coupling are a convolution of the quantum states with the exciting electromagnetic field. Succeeding in confining the optical excitation to a very small volume below the diffraction limit, implies the presence of optical fields with high lateral spatial frequencies, determining coupling matrix elements that can differ significantly from far-field ones. The most striking manifestation of these effects is the breaking of the usual optical selection rules.
and the possibility to excite dark states whose optical excitation is forbidden by symmetry in the far field. Moreover, dark states are not able to emit light in the far-field, for they generate only fields with high in-plane wavevectors corresponding to evanescent waves. Hence their lifetimes at low temperatures result significantly increased with respect to their bright counterparts.

In Part III we provided a general near-field theoretical model for near-field optical spectroscopy. We calculated in eq. (8.32) the time evolution for the most generic quantity to be addressed in order to study the optical response under coherent excitation of a generic QW in the weak-coupling case. By means of the DCTS we have been able to give at any order of the coherent exciting field the system of first order differential equations to be solved numerically in order to gather the output optical field witnessing the nonlinear dynamics inside the heterostructure. The first case-study analyzed has been near-field spectroscopy of QW with interface disorder, i.e. naturally occurring quantum dots. More specifically, we have investigated the impact of the reduced dark-states relaxation on the distribution of electron-hole pairs among the available energy levels after a far-field continuous wave excitation resonant with the barrier energy levels. At low temperatures all the dot level-occupations but one decrease monotonically with energy. The uncovered exception, corresponding to the second (dark) energy level, displays a steady-state carrier density exceeding that of the lowest level more than a factor two. The root cause is not radiative recombination before relaxation to lower energy levels, but at the opposite, carrier trapping due to the symmetry-induced suppression of radiative recombination. Such a behaviour can be observed by collection-mode near-field optical microscopy. The calculated near-field luminescence properties of these states depend critically on tip position, temperature and spatial resolution and clearly indicate the potentiality of near-field PL for addressing general questions regarding nanoscale energy transfer in open nanosystems.
Appendix A

Damping terms in Markov approximation

The acoustic phonon interaction Hamiltonian in chapter 8 consider the case of a quantum well without disorder, indeed already in the Hamiltonian we have explicitly taken into account the wavevector conservation rule:

\[
\hat{H}_{ep} = \sum_{Nk;\zeta Nq;\eta} t^q_{Nk;\zeta Nq;\eta} \hat{X}_{Nk+q;Nq} (\hat{b}_{q;\eta} + \hat{b}^\dagger_{-q;\eta}) ,
\]

(A.1)

where \(\zeta, \eta\) are the proper quantum number spanning the associated N-pair state space (e.g. if \(N = 1\) \(\zeta = (n, \sigma)\)). All the model ingredients are clumped into \(t\):

\[
t^q_{Nk;\zeta Nq;\eta} = \sum_{k} \left( \frac{\hbar|q|}{2\rho v_{s} v} \right)^{1/2} \left( \langle N\zeta(k+q) | D_{\zeta} a_{k+q}^{\dagger} \hat{c}_{k} + D_{\eta} a_{k+q}^{\dagger} \hat{d}_{k} | N\eta k \rangle \right) .
\]

(A.2)

In this appendix we will give the generic Markov calculation for the damping in this situation as a sort of reference for possible future implementations.

acoustic phonons

This form is a little bit different from (2.87) and it leads to a compact form similar to eqs. (2.100) and (2.101). It has been already used in Chapter 5.1 in (5.18), we
will rewrite it in the following for reference:

\[
\frac{d}{dt} \langle \hat{O}(t) \rangle_S = -\frac{1}{\hbar^2} \sum_{q, \pm} \int_0^\infty du \left( n_{\pm q}^R + \theta(\pm) \right) \times \left( e^{i \frac{\vec{R} \cdot \vec{u}}{\hbar}} \langle [\hat{O}, \hat{\mathcal{Q}}_q] \hat{\mathcal{Q}}_{-q}(-u) \rangle_S - e^{-i \frac{\vec{R} \cdot \vec{u}}{\hbar}} \langle \hat{Q}_{-q}(-u) [\hat{O}, \hat{\mathcal{Q}}_q] \rangle_S \right) \tag{A.3}
\]

For the generic operator \( \hat{O} = \hat{X}_{\alpha k; \beta k} \), the calculation follows

\[
\hat{Q}_q = \sum_{Nk; \eta} t_q^{Nk; \eta} | N\zeta(k + q^\parallel) \rangle \langle N\eta k | , \tag{A.4}
\]

\[
\frac{d}{dt} \langle \hat{X}_{\alpha k; \beta k} \rangle_{H_{\text{ep}}} = -\frac{1}{\hbar^2} \sum_{q, \pm} \int_0^\infty du \left( n_{\pm q}^R + \theta(\pm) \right) \left[ e^{i \frac{\vec{R} \cdot \vec{u}}{\hbar}} \left( \sum_{\eta, \eta} e^{-i(\omega M\eta(k\beta - q) - \omega M\eta q_k)} t_{Nk; \eta}^{\alpha q} t_{\eta M\eta k; \eta}^{\beta q} \langle \hat{X}_{\alpha k; \beta k} \rangle_{H_{\text{ep}}} - \sum_{\eta, \eta} e^{-i(\omega M\eta(k\beta + q) - \omega M\eta q_k)} t_{Nk; \eta}^{\alpha q} t_{\eta M\eta k; \eta}^{\beta q} \langle \hat{X}_{\alpha k; \beta k} \rangle_{H_{\text{ep}}} \right) \right] - \sum_{\zeta, \zeta} e^{i \frac{\vec{R} \cdot \vec{u}}{\hbar}} \left( \sum_{\eta, \eta} e^{-i(\omega N\zeta(k\alpha - q) - \omega N\eta q_k)} t_{Nk; \eta}^{\alpha q} t_{\eta Nk; \eta}^{\alpha q} \langle \hat{X}_{\alpha k; \beta k} \rangle_{H_{\text{ep}}} - \sum_{\eta, \eta} e^{i(\omega N\zeta(k\alpha + q) - \omega N\eta q_k)} t_{Nk; \eta}^{\alpha q} t_{\eta Nk; \eta}^{\alpha q} \langle \hat{X}_{\alpha k; \beta k} \rangle_{H_{\text{ep}}} \right) \tag{A.5}
\]

we shall retain only the so-called T1 and T2, then

\[174\]
$$= -\frac{1}{\hbar} \sum_{\mathbf{q}, \pm} \left( n^R_{\mathbf{q}} + \theta(\pm) \right) \pi$$

$$\left[ \delta_M \sum_{\eta} \left( \hbar \omega_{M\eta} (k_\beta - \mathbf{q}) - \hbar \omega_{M\beta} k_\beta \pm \epsilon^R_{\mathbf{q}} \right) \left| t^q_{M(k_\beta - \mathbf{q}); \beta \eta} \right|^2 \left\langle \hat{X}_{N\alpha k_\alpha; M\beta k_\beta} \right\rangle -$$

$$- \delta_M \delta_{\alpha, \beta} \delta_{k_\alpha, k_\beta} \sum_{\xi} \delta (\hbar \omega_{M\xi} k_\beta - \hbar \omega_{M\xi(k_\beta + \mathbf{q})} \pm \epsilon^R_{\mathbf{q}}) \left| \sum_{\eta} t^q_{N(k_\alpha - \mathbf{q}); \alpha \eta} \right|^2 \left\langle \hat{X}_{N\xi(k_\alpha + \mathbf{q}); N\xi(\xi \beta \eta)} \right\rangle \right) -$$

$$\delta_M \delta_{\alpha, \beta} \delta_{k_\alpha, k_\beta} \sum_{\eta} \delta (\hbar \omega_{N\alpha k_\alpha} - \hbar \omega_{N\eta(k_\alpha - \mathbf{q})} \pm \epsilon^R_{\mathbf{q}}) \left| t^q_{N(k_\alpha - \mathbf{q}); \alpha \eta} \right|^2 \left\langle \hat{X}_{N\xi(k_\alpha - \mathbf{q}); N\xi(\xi \beta \eta)} \right\rangle +$$

$$+ \sum_{\xi} \delta (\hbar \omega_{N\xi(k_\alpha + \mathbf{q})} - \hbar \omega_{N\alpha k_\alpha} \pm \epsilon^R_{\mathbf{q}}) \left| t^q_{N(k_\alpha + \mathbf{q}); \alpha \eta} \right|^2 \left\langle \hat{X}_{N\xi(k_\alpha + \mathbf{q}); N\xi(\xi \beta \eta)} \right\rangle \right] \right) \tag{A.6}$$

Manipulating further, changing $\mathbf{q} \to -\mathbf{q}$, resumming in reverse order, with the properties $t^q_{M(k_\beta - \mathbf{q}); \beta \eta} = (t^q_{(k_\beta - \mathbf{q}); \beta \eta})^*$, $n^R_{-\mathbf{q}} = n^R_{\mathbf{q}}$, $\omega^R_{-\mathbf{q}} = \omega^R_{\mathbf{q}}$, where needed,

$$\frac{d}{dt} \left\langle \hat{X}_{N\alpha k_\alpha; M\beta k_\beta} \right\rangle_{\text{ep}} = -\frac{1}{2} \left( \Gamma^N_{\alpha, k_\alpha} + \Gamma^M_{\beta, k_\beta} \right) \left\langle \hat{X}_{N\alpha k_\alpha; M\beta k_\beta} \right\rangle +$$

$$+ \frac{1}{2} \delta_{M, N} \delta_{\alpha, \beta} \delta_{k_\alpha, k_\beta} \sum_{\xi k'} \left( W^{(N)}_{\alpha k_\alpha, \xi k'} + W^{(M)}_{\beta k_\beta, \xi k'} \right) \left\langle \hat{X}_{N\xi k'; N\xi k'} \right\rangle \tag{A.7}$$

with the usual scattering rates, in the following they are written already taken into account that we are into a QW thus the Npair $k$’s are two dimensional whereas the phonon has three dimensions, we shall consider as usual the confined direction as $z$

$$W^{(N)}_{\eta k', \alpha k_\alpha} = \frac{2\pi}{\hbar} \sum_{q_z} \left| \sum_{\eta k_{\alpha, q_z}} \left( \hbar \omega_{N\eta k'} - \hbar \omega_{N\eta k_{\alpha, q_z}} \pm \epsilon^R_{(k'_{\alpha, q_z})} \right) \left( n^R_{(k'_{\alpha, q_z})} + \theta(\pm) \right) \right|^2$$

$$W^{(N)}_{\eta k', \alpha k_\alpha} = \sum_{\pm} W^{(N)}_{\eta k', \alpha k_\alpha}$$

$$\Gamma^N_{\alpha, k_\alpha} = \sum_{\xi k'} W^{(N)}_{\eta k', \alpha k_\alpha} \tag{A.8}$$

**Radiative scattering**

In this case no particular symmetry property of the operators in the interaction Hamiltonian can be exploited and the most general form of eq. (2.100) has to be
used. It reads

\[
\frac{d}{dt} \langle \hat{\mathcal{O}} \rangle(t) = \langle \left[ \hat{\mathcal{O}}, \hat{H}_0 \right] \rangle_S - \frac{1}{\hbar^2} \sum_i \int_0^\infty du \left\{ \right.
\]

\[
\times \left[ (n_i^R + g_i) \left( e^{-\frac{\hbar \mathcal{O}(u)}{\hbar}} \langle \hat{\mathcal{O}} \hat{Q}_i^\dagger \rangle_S - e^{-\frac{\hbar \mathcal{O}(u)}{\hbar}} \langle \hat{Q}_i \hat{\mathcal{O}}^\dagger \rangle_S \right) +
\]

\[
+ n_i^R \left( e^{-\frac{\hbar \mathcal{O}(u)}{\hbar}} \langle \left[ \hat{\mathcal{O}}, \hat{Q}_i \right] \hat{Q}_i^\dagger \rangle_S - e^{-\frac{\hbar \mathcal{O}(u)}{\hbar}} \langle \left[ \hat{Q}_i, \hat{\mathcal{O}} \right] \hat{Q}_i \rangle_S \right) \right\}
\]

(A.9)

Again for the generic operator \( \hat{\mathcal{O}} = \hat{X}_{N\alpha, \beta k} \) one obtains

\[
\hat{Q}_i = \hat{P}^{- \sigma}_k, \quad \hat{F}_i = \hat{E}^{+ \sigma}_k
\]

(A.10)

and

\[
\frac{d}{dt} \langle \hat{X}_{N\alpha, \beta k} \rangle |_{\hat{\theta}^L_M} = -\frac{1}{\hbar^2} V \sum_{\sigma k} \sum_{n\eta} \frac{M_{n\sigma} M_{\eta k}^*}{V} \int_0^\infty du \left[ \right.
\]

\[
e^{-i \frac{\hbar \mathcal{O}_k}{\hbar}} (n_{\sigma_k} + g_{\sigma_k}) \sum_{\rho k_{\rho}, \mu k_{\mu}} e^{-i (\omega_{M-1} \rho, \mu)_{k_{\rho}, k_{\mu}}} \langle \mathcal{B}_{n\sigma_k} | (M-1) \rho k_{\rho} \rangle \langle (M-1) \rho k_{\rho} | \mathcal{B}_{n\sigma_k} \rangle -
\]

\[
e^{-i (\omega_{M} \rho, \mu)_{k_{\rho}, k_{\mu}}} \langle \mathcal{B}_{n\sigma_k} | (M+1) \mu k_{\mu} \rangle \langle (M+1) \mu k_{\mu} | \mathcal{B}_{n\sigma_k} \rangle -
\]

\[
\left. \right. \left. \right\}
\]

176
\[ -e^{-i(\omega \eta \kappa - \omega_{N+1}\rho \kappa_{\rho})/\hbar} \langle N\eta \kappa | \hat{B}_{\pi\sigma k} | (N+1)\rho \kappa_{\rho} \rangle \\
\langle (N+1)\rho \kappa_{\rho} | \hat{B}^\dagger_{\pi\sigma k} | N\alpha \kappa_{\alpha} \rangle \langle \hat{X}_{N\eta \kappa_{\eta}; M\beta \kappa_{\beta}} \rangle - \\
\langle (N+1)\rho \kappa_{\rho} | \hat{B}^\dagger_{\pi\sigma k} | N\alpha \kappa_{\alpha} \rangle \langle \hat{X}_{N\eta \kappa_{\eta}; M\beta \kappa_{\beta}} \rangle - \]
\[ e^{-i\frac{\hbar}{\hbar} n_{\pi k}} \sum_{\eta \kappa_{\alpha}, \rho \kappa_{\rho}} \left( e^{-i(\omega_{M+1}\eta \kappa_{\eta} - \omega_{M\rho \kappa_{\rho}})/\hbar} \right) \\
\langle M\beta \kappa_{\beta} | \hat{B}_{\pi\sigma k} | (M+1)\eta \kappa_{\eta} \rangle \langle (M+1)\eta \kappa_{\eta} | \hat{B}^\dagger_{\pi\sigma k} | M\rho \kappa_{\rho} \rangle \langle \hat{X}_{N\alpha \kappa_{\alpha}; M\rho \kappa_{\rho}} \rangle - \\
\langle (N+1)\eta \kappa_{\eta} | \hat{B}_{\pi\sigma k} | M\rho \kappa_{\rho} \rangle \langle \hat{X}_{N\alpha \kappa_{\alpha}; M\rho \kappa_{\rho}} \rangle \left( \langle (N+1)\eta \kappa_{\eta} | \hat{B}_{\pi\sigma k} | (N-1)\eta \kappa_{\eta} \rangle \left( \langle N\eta \kappa_{\eta} | \hat{B}^\dagger_{\pi\sigma k} | (N+1)\eta \kappa_{\eta} \rangle - \\
\langle (N-1)\eta \kappa_{\eta} | \hat{B}_{\pi\sigma k} | N\alpha \kappa_{\alpha} \rangle \langle \hat{X}_{N\kappa_{\kappa}; M\beta \kappa_{\beta}} \rangle \right) \right), \\
\]

retaining the T1 and T2 approximations we obtain a \( \delta_{\eta,\pi} \) because the same state transition must be connected by the same interband transition, i.e. the same exciton operator. Eventually we are left with

\[ \frac{d}{dt} \langle \hat{X}_{N\alpha \kappa_{\alpha}; M\beta \kappa_{\beta}} | \hat{H}^M_{\pi} \rangle = -\frac{1}{2} \left( \Gamma^\dagger_{\alpha \kappa_{\alpha}} + \Gamma^\dagger_{\beta \kappa_{\beta}} \right) \langle \hat{X}_{\alpha \kappa_{\alpha}; \beta \kappa_{\beta}} \rangle + \\
\delta_{N,M} \delta_{\alpha,\beta} \delta_{\kappa_{\alpha}, \kappa_{\beta}} \sum_{\rho \kappa_{\rho}} \left[ W^{(-)l}_{N\alpha \kappa_{\alpha}; (N+1)\rho \kappa_{\rho}} \langle \hat{X}_{(N+1)\rho \kappa_{\rho}; (N+1)\rho \kappa_{\rho}} \rangle + \\
+ W^{(+l)}_{N\alpha \kappa_{\alpha}; (N-1)\rho \kappa_{\rho}} \langle \hat{X}_{(N-1)\rho \kappa_{\rho}; (N-1)\rho \kappa_{\rho}} \rangle \right], \]  

(A.11)

where

177
\begin{align*}
W_{Rf_{k_f};P_{k_i}}^{(\pm)} &= \frac{2\pi}{\hbar} \sum_{k_{\sigma\tau}} |M_{n_{\sigma\tau}}|^2 \left[ n_{k_{\sigma\tau}} \right] \delta(\hbar\omega_{Rf_{k_f}} - \omega_{P_{k_i}} - \hbar\omega_{k_{\sigma\tau}})|\langle Rf_{k_f} | \hat{B}_{n_{\sigma\tau}} | P_{k_i} \rangle|^2 \\
W_{Rf_{k_f};P_{k_i}}^{(\mp)} &= \frac{2\pi}{\hbar} \sum_{k_{\sigma\tau}} |M_{n_{\sigma\tau}}|^2 \left[ n_{k_{\sigma\tau}} + g_{k} \right] \delta(\hbar\omega_{Rf_{k_f}} - \omega_{P_{k_i}} + \hbar\omega_{k_{\sigma\tau}})|\langle Rf_{k_f} | \hat{B}_{n_{\sigma\tau}} | P_{k_i} \rangle|^2 \\
\Gamma_{Qq_{k_i}} &= \sum_{\rho \neq \sigma \pm} W_{(Q\pm1)(\rho\sigma),Qq_{k_i}}^{(\pm)} \\
\text{(A.12)}
\end{align*}
Appendix B

Schrödinger equation for a generic N-pair aggregate

Let us consider a generic heterostructure where it is possible to separate the generic coordinate \( r \) into a free \( x \) and a confined direction \( z \), thus \( r = (x,z) \). In the specific problem of a QW we have \( r = (x,z) \), where the first is a two-dimensional vector whereas we face a single confining direction usually denoted by \( z \).

The generic Schrödinger equation for a complex made up of \( k \) constituent is

\[
\sum_a \left[ -\frac{\hbar^2}{2m_a} \nabla_a^2 (r_a) + V_a(r_a) + \sum_{b \neq a} U_{ab} \right] \Psi(\{r\}_{i=1}^k) = E^{3D} \Psi(\{r\}_{i=1}^k). \tag{B.1}
\]

The separation into free and confined direction results in a factorized form of the wavefunction:

\[
\Psi(\{r\}_{i=1}^k) = \Psi^{2D}(\{x\}_{i=1}^k) \prod_a \chi_a(z_a), \tag{B.2}
\]

where \( a \) runs over all particles (e,h for the exciton and e1,e2,h1,h2 for the biexciton) and \( \chi_a(z_a) \)'s are the singleparticle subband functions. The resulting in-plane effective mass Schrödinger equation for the exciton and biexciton reads as
\[
\sum_a \left[-\frac{\hbar^2}{2m_a} \nabla_a^2 (x_a) + \tilde{V}_a(x_a) + \sum_{b \neq a} \tilde{U}_{ab}\right] \Psi^{2D}(\{x\}) = \left( E^{3D} - \sum_a E_z^a \right) \Psi^{2D}(\{x\}),
\]  

(B.3)

In reasonable good quality quantum structures the amplitude of the confinement energy fluctuations is typically 1 order of magnitude smaller than the exciton binding energy [129]. In this limit, the relative exciton motion may be assumed to be undistorted by disorder. Disorder significantly affects only the COM motion through an effective potential \(\tilde{V}_a(R)\), where \(R\) now indicates the COM vector whose coordinates are given by

\[
R = \frac{1}{M} \sum_a m_a x_a,
\]

(B.5)

where \(M = \sum_a m_a\) is the complex kinetic mass, and

\[
q_{ij} = x_j - x_i
\]

(B.6)

is the generic relative coordinate. In the exciton case the complex is formed by two constituent, one electron with \(a = e\) and a hole \(a = h\), thus we have a single relative coordinate say \(\rho = x_e - x_h\) and the customary situation is recovered. On the contrary when dealing with at least a biexciton, we have 6 relative coordinate out of which only 3 of them are linearly independent, it will be a matter of convenience to choose the relative set (we shall symbolically denoting it as \(\{q\}\)).
The wave function which factorized in relative motion and COM are a complete set, as a consequence we can expand the solution of the Schrödinger equation as

\[ \Psi^{2D}(\{x\}) = \sum_{n,\mu,a} \Psi_{\alpha,\mu}(R)\phi_{n,\mu}(\{q\}) , \]  

or in Dirac notation

\[ \langle r,\{q\}|\Psi^{2D}\rangle = \sum_{n,\mu,a} \langle R|\alpha\mu\rangle\langle\{q\}|n\mu\rangle , \]

we shall consider the very generic case where there could be independent \((\alpha,n)\) as well as shared \((\mu)\) quantum indexes. In matrix form eq. (B.3) is an eigenvalue problem

\[ \sum_{\mu\alpha n} \left( \langle \mu' n' | \langle \mu' \alpha' | H^{2D} | \mu \alpha \rangle - E^{2D} \delta_{\mu' n' \alpha' \mu n} \right) \phi_{n,\mu}(\{q\}) = 0 , \]

where

\[ H^{2D}(R,\{q\}) = -\frac{\hbar^2}{2M} \nabla_R^2 - \frac{\hbar^2}{2} \sum_{a,b} \frac{1}{m_a} \frac{d^2}{dq_{a,b}^2} + \sum_{a,b} \tilde{U}_{ab}(q_{a,b}) + \sum_a \tilde{V}_a(R,\{q\}) \]  

By definition the wave functions describing the relative motion are the solution of the Schrödinger equation

\[ \left[ -\frac{\hbar^2}{2} \sum_{a,b} \frac{1}{m_a} \frac{d^2}{dq_{a,b}^2} + \sum_{a,b} \tilde{U}_{ab}(q_{a,b}) \right] \phi_{n,\mu}(\{q\}) = \hbar \omega_{n,\mu} \phi_{n,\mu}(\{q\}) . \]

Under the hypothesis that we shall know these relative wave function we can plug eq. (B.11) into eq. (B.9) projecting onto the COM subspace.

\[ \sum_{\mu\alpha n} \left( \langle \mu' n' | \langle \mu' \alpha' | T_{COM}^{2D} + \sum_a \tilde{V}_a(R,\{q\}) \rangle | \mu \alpha \rangle - E^{2D} - \hbar \omega_{n,\mu} \right) \delta_{\mu' n' \alpha' \mu n} = 0 , \]
on the COM space only it reads

$$
\sum_{\mu,\alpha} \left[ T_{\mu'\alpha'}^{\text{COM}} \delta_{\mu'\alpha';\mu\alpha} + \langle \mu'\alpha' | \left( \sum_{n',n} \langle \mu' n' | \sum_{a} \hat{V}_a(R_n,\{q\}) | \mu n \rangle \right) | \mu\alpha \rangle \right] = (B.13)
$$

\[= \left( E^{2D} - \sum_n \hbar \omega_{n,\mu} \right) | \mu,\alpha \rangle. \]

In real-space coordinate it has the highly evocative form

$$
\sum_{\mu,\alpha} \left( - \frac{\hbar^2}{2M} \nabla^2_{R_{\mu'\mu}} + V_{\mu'\mu}(R) \right) \Psi_{\alpha,\mu}(R) = \left( E^{2D} - \sum_n \hbar \omega_{n,\mu} \right) \Psi_{\alpha,\mu}(R), \quad (B.14)
$$

where

$$
V_{\mu'\mu}(R) = \sum_{n,n'} \int d\{q\} \phi_{n',\mu'}^*(\{q\}) \left( \sum_{a} \hat{V}_a(R_n,\{q\}) \right) \phi_{n,\mu}(\{q\}) \quad (B.15)
$$
Bibliography


Bibliography


[113] J. S. Bell, Physics (Long Island City, N.Y.) 1, 195 (1964).


