Stochastic dynamics of microcavity polaritons

A.E. Pedraza *, L. Quiroga

Departamento de Física, Universidad de Los Andes, A.A. 4976, Bogotá D.C., Colombia

Abstract
We study the time dependent polariton condensation as well as the parametric scattering process of polaritons in a semiconductor microcavity. Based upon a new stochastic scheme the dynamics for both cases is fully analyzed. We show how the evolution of the system is described by a set of stochastic differential Schrödinger equations which in average reproduces the exact dynamics. Furthermore, we underline the role that Coulomb correlations plays in the polariton dynamics. Threshold behaviors are well captured by the present approach. The results are in complete agreement with recent experimental observations.

Key words: D. Bose-Einstein condensation, D. microcavity polaritons.
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1. Introduction

Microcavity polaritons are quasiparticles created in the strong coupling regime between photons in a microcavity and the excitonic resonance of a semiconductor quantum well [1]. Polaritons satisfy Bose statistics (at least in the low density regime) and are excellent candidates to study quantum effects at the macroscopic level, e.g. Bose-Einstein Condensation (BEC) and superfluidity. As a practical advantage that polariton systems exhibit over other quasi-bosonic particles in condensed matter systems is their easy experimental optical control [2,3]. Particulary, polaritons can be optically injected in the microcavity by external light impinging on the semiconductor nanostructure, and their properties can be inferred from the observation of the emitted light. The possibility of obtaining a quantum fluid in a solid state system, with easy control and integration potentialities, could also open a new promising way to the implementation of quantum information technologies.

The polariton condensation has being actively pursued, since no BEC has being observed for a solid state system. Recently, a possible quantum phase transition of polaritons was suggested [4], and the spontaneous formation of a coherent polariton state was reported in [5]. This last result shows a remarkable manifestation of the bosonic behavior of this short living particles, but it has been interpreted as a polariton laser phase transition. From the theoretical side, most existing works [4] pursue a strict analogy with the laser theory, where the quantum fluctuations responsible for the phase transition appear directly from the particle number conservation.

On the other hand, it is well known that due to their excitonic component, polaritons are subjected to Coulomb interactions. This nonlinearity is expected to trigger the quantum phase transition. Moreover, the unexpected observation of a thermal-like intensity far above the threshold [5] does not match with the laser picture, leaving the unanswered question of the role of interactions, usually overlooked in the theory. A more recent attempt to address this question has been made in [6], where the laser picture was left behind and polariton-polariton interactions were taken into account. Nonetheless, this last study is within the scope of a mean-field approximation (MFA), and thus fails to give a proper description of the polariton dynamics above the parametric threshold where high order Coulomb correlations are more important than in the low density regime.

The present paper is devoted to gain more physical insight into the polariton dynamics by analyzing two different dynamical processes: (i) the time-dependent condensation and coherence build-up of polaritons relaxing into the lowest branch states and (ii) the time-dependent parametric scattering. Both processes will be considered within the same theoretical approach based on the stochastic unravelling of the dynamics of an interacting many-body system embedded in a noisy environment. Partial realizations of such a computational scheme have been previously consid-
ered. On the one hand, closed quantum interacting systems have been studied by stochastic unravellings in Ref.[7] for bosons and in Ref.[8] for fermions. On the other hand, simple open quantum systems have been studied by following the stochastic density matrix for the whole system plus environment in Refs.[10,11]. It is one of the main aims of the present work to extend those partial developments to a complex many-body open quantum system such as the one offered by microcavity polaritons. In order to provide quantitative predictions, previous stochastic schemes are extended to follow the evolution of a polariton system coupled to a noisy environment where interparticle interactions and environment effects can be treated on the same footing. This will allow us to transform the evolution of an open interacting polariton system, into the stochastic evolution of a single polariton [7] separately from the stochastic evolution of the environment [10,11]. As compared with similar approaches the advantages of this stochastic scheme are manifold: first of all, the complete dynamics, both transient and steady-state regimes, can be obtained with full fluctuations due to interactions properly taken into account. Second, a many-polariton problem can be mapped onto a single polariton system driven by stochastic terms.

The paper is organized as follows: in Section 2 a general description of the stochastic scheme is presented. In Section 3 the polariton condensation dynamics with both Coulomb interactions and phonon scattering is considered. We pay particular attention to the quantum properties of the condensate fraction and the second-order degree of coherence. The parametric scattering of pump polaritons in the lower polariton branch and the dynamics of formation of the eight-shaped final allowed states is studied in Section 4, where comparison to experimental results [2] reveals the importance of including the nonlinearities associated to polariton interactions, specially above the threshold regime. Finally, in Section 5 we sketch our concluding remarks.

2. Stochastic scheme for interacting bosons

We start with a brief review of the main assumptions for a stochastic scheme aiming to determine the evolution of a closed many-boson system [7]. The initial full information of the system is encoded in the state vector $|\psi\rangle = |N : \phi\rangle$ where N bosons are accommodated in the same single particle state $|\phi\rangle$. Thus, one needs only to find the appropriate evolution of $|\phi\rangle$ in order to obtain the N-bosons system dynamics. For non interacting particles this is of course trivial, but in the interacting case the action of the Hamiltonian on the N-particle state vector produces terms orthogonal to $|\phi\rangle$. This cannot be taken into account by standard MFA, and hence the necessity to introduce a noise term with specific properties. The first non-trivial application of this method was performed in Ref.[12].

For microcavity polariton systems at low temperatures, an always present source of nonlinearity is the coupling with the acoustic phonon reservoir. At first glance, the necessity to include this reservoir, should leave the stochastic scheme just mentioned out of the picture for polariton dynamics. However, as it has recently been stated for simple open systems [10,11], the quantum system and its environment can be described by a pair of decoupled Stochastic Schrodinger Differential Equations (SSDE). The success of this fictitious evolution is based on the adequate introduction of stochastic terms. This will guarantee that when averaged over the stochastic trajectories of both subsystems, the exact whole dynamics is recovered. The central quantum system, however decoupled from its surrounding in every individual trajectory, will feel the effect of the environment through a driven noise term, allowing in this way to obtain the exact reduced density matrix at any time. In the following of this Section we will give a brief guideline on how the two just mentioned stochastic schemes can be extended to consider polariton dynamics.

In general, the full Hamiltonian can be written as

$$H_T = H_S + H_E + H_i$$

(1)

where $H_S$ describes the central quantum subsystem (including interparticle interactions), $H_E$ is the Hamiltonian associated to the environment and $H_i$ is the coupling Hamiltonian which we assume can be expressed as $H_i = \sum_\alpha A_\alpha \otimes B_\alpha$, where $A$ and $B$ are operators acting on the system and environment, respectively. In order to obtain the formally exact density matrix as the stochastic average over multiple realizations, each sample has to be taken as the dyadics $\rho_i = |\Phi_1\rangle\langle\Phi_2|$. Moreover, one of the main advantages of the present approach is that the different kets and bras in the dyadics can be taken as the separable forms $|\Phi_\nu\rangle = |\psi_\nu\rangle \otimes |\chi_\nu\rangle$ ($\nu = 1,2$) of system and environment state vectors, respectively. Of course, the stochastic average reproduces the true entanglement between quantum system and environment, as it should be. The evolution of the whole system under the Hamiltonian in Eq.(1) is then given by the set of SSDEs [10]

\[d|\psi_\nu(t)\rangle = -idt H_S|\psi_\nu(t)\rangle - idt \sum_\alpha dW_{\alpha\nu}(t)A_\alpha |\psi_\nu(t)\rangle\]

(2)

\[d|\chi_\nu(t)\rangle = -idt H_E|\chi_\nu(t)\rangle + \sum_\alpha dW_{\alpha\nu}(t)B_\alpha |\chi_\nu(t)\rangle\]

(3)

with noise terms satisfying $E[dW_{\alpha\nu}(t)dW_{\alpha'\nu'}(t)] = \delta_{\alpha,\alpha'}\delta_{\nu,\nu'}$ and $E[dW_{\alpha\nu}] = 0$ ($E[...]$ means the average over stochastic realizations). By averaging the dyadics built on the states evolving as in Eq.(2) and Eq.(3), the exact Liouville equation for the whole density operator is found. This has been extensively discussed in Ref.[10] within a quantum jump approach and alternatively in Ref.[11] using a diffusion method.

Assuming the Hamiltonian $H_S$ in Eq.(1) conserves the number of particles, one can consider the dynamics of a closed quantum system due to the separability from the environment. Hence, the new idea is to describe the evolution of the N interacting particles, now in an open situation, as
occupying the single particle state vector $|\phi\rangle$. In order to find the stochastic evolution within this prescription, the new form of the state vector $|\psi\rangle = |N: \phi\rangle$ has to be adapted to Eq.(2), verifying that the SSDEs once again yield to the formally exact dynamics. This procedure implies that

$$d|\phi_{\nu}\rangle = -idtG_{\nu}|\phi_{\nu}\rangle + dM_{\nu}|\phi_{\nu}\rangle - i\sum_\alpha \tilde{a}_\alpha|\phi_{\nu}\rangle dW_{\alpha\nu}(t) \tag{4}$$

where $G_{\nu}$ contains inter-particle interaction effects treated at the level of MFA while $dM_{\nu}$ is a noise term responsible for capturing the particle-particle interaction effects beyond MFA. Finally, the third term in Eq.(4) carries the net effect of the environment coupled to the system. The notation $\tilde{a}_\alpha$ is used to represent a system’s coupling operator projected into the single particle space.

It is important to note that within this stochastic approach, where system and environment are always separable for each individual realization, the calculation of any system’s observable, after tracing over the environment, is given by

$$\langle \hat{O}(t) \rangle = \langle \langle \psi_2(t) | \hat{O}(t) | \psi_1(t) \rangle \langle \chi_2 | \chi_1 \rangle \rangle. \tag{5}$$

With this in mind, a complex problem where bosonic particles interact with each other and with their surroundings, has been reduced to a set of SSDEs for a single particle, Eq.(4), decoupled from its environment, Eq.(3). The price to be paid for this formidable reduction of complexity is that a large number of stochastic realizations has to be performed in order to recover the exact solution. Obviously, the results obtained by this procedure are beyond MFA while additionally particle-particle interactions as well as environment effects (phonons) can be treated on equal grounds.

### 3. Spontaneous coherence buildup of microcavity polaritons

In a typical photoluminescence experiment under non-resonant excitation, the strong energy dispersion of microcavity polaritons leads to a bottleneck relaxation dynamics [14], where polaritons go to the flat exciton-like region of the energy dispersion curve. From this point on, polaritons relax into the bottom states of the lower branch. Using these considerations a simplified model of polariton dynamics is an effective two-level system [3], where the ground state corresponds to $k = 0$ which is non-degenerate ($E_0$), and the excited or bottleneck set of states is macroscopically degenerate ($E_1$) (see Figure 1).

In the lower polartion branch the Hamiltonian describing both Coulomb and phonon scattering is

$$H = \sum_k \omega_k p_k^{\dagger} p_k + \sum_q \omega_q b_q^{\dagger} b_q + H_C + H_{ph} \tag{6}$$

$$H_C = \frac{1}{2} \sum_{kk'} q_{kk'}(q) p_{k+q}^{\dagger} p_{k'} - q p_{k'} p_k \tag{7}$$

$$H_{ph} = \sum_{kk'} g_{kk'}(q) (b_q^{\dagger} p_k^{\dagger} + H.C.) \tag{8}$$

where $p_k$ is the Bose annihilation operator for a polariton with momentum $k$. The excitonic component of the polaritons is responsible for Coulomb interactions, which is captured in $H_C$ with the coupling strength $v_{kk'}^q$. The semiconductor environment is modelled by acoustical phonons distributed according to the lattice temperature. The corresponding phonon annihilation operator is $b_q$ with the usual Bose commutation relations $[b_q, b_{q'}^\dagger] = \delta_{qq'}$. The polariton-phonon scattering is responsible for decoherence and is represented in Eq.(6) as $H_{ph}$ with $g_{kk'}^qdq$ as the coupling strength.

Note that this polariton Hamiltonian is number conserving. The dynamical condensation of microcavity polaritons will be studied using the stochastic formalism exposed in the latter Section. The relevant material parameters [2] are typical for a GaAs-based microcavity: heavy-hole exciton Rabi splitting $\hbar \Omega_R = 1.82 meV$, the detuning with the confined photon mode $-2.4meV$ and the effective index of the cavity mode 3.5. The quantization area $A = 100 \mu m^2$ determines the Coulomb interaction [13] and the phonon coupling terms [14].

We consider $N$ interacting polaritons in the presence of a semiconductor environment at zero temperature. Since, the effective two level model does not consider the relaxation from high energy states into the effective bottleneck state, the initial state consists of the total number of polaritons randomly distributed in the excited energy level (importantly, no seed is needed for level $E_0$). Indeed, a zero temperature environment will force the polaritons into their ground state following a non-trivial evolution due to polariton interactions. Our main interest is to give a full description of the ground state statistics. We first calculate the ground state probability distribution $P_0(n, t)$. This is initially a delta function centered at $n = 0$, since all polaritons are initially in the excited state. For comparison purposes, an overall factor, $\gamma$, is introduced in the Coulomb interaction term in order to account for the effective strength of two body Coulomb interactions: $\gamma = 0$ corresponds to no Coulomb interaction. As is clearly shown in Figure 2, for a fixed total number of polaritons $N = 1600$, the polariton system decays monotonically into its ground state

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**Fig. 1.** Lower polariton energy branch versus total momentum in a linear-log scale. The excited state is represented by the effective bottleneck state, which is at the edge of the flat exciton-like dispersion region. The ground state is the zero momentum state in the bottom of the dispersion curve.
and (d) \( \gamma = 1 \), averaged over \( 5 \times 10^5 \) realizations. The total number of polaritons is \( N = 1600 \) and lattice temperature \( T=0 \).

Fig. 3. Projections of the ground state probability distribution \( P_0(n,t) \) at four various times: 100 ps (green), 200 ps (red), 300 ps (blue) and 400 ps (purple). Results are shown for \( N=1600 \), \( 5 \times 10^5 \) realizations and different polariton interaction strengths: (a) \( \gamma = 0 \), (b) \( \gamma = 0.4 \), (c) \( \gamma = 0.8 \) and (d) \( \gamma = 1 \).

when interactions are not included. As the interactions are turned on, the system’s dynamics changes completely. Polaritons start with a slow relaxation dynamics, but after a finite time the ground state becomes rapidly populated with a macroscopic fraction of \( N \).

The Gaussian-like distribution for vanishing inter-particle interactions is evidently different from the Poisson-like distribution when the interactions are fully included, as displayed in Fig. 3. This latter feature is a clear signature of an interaction driven coherence buildup process for which an off-diagonal-long-range order (ODLRO) can develop. It is well known [15] that ODLRO does not implies a finite value of \( \langle p_0 \rangle \), in agreement with our results.

The so-called coherence degree parameter is defined as \( \eta = 2 - g^{(2)}(0) \), with \( g^{(2)}(0) = \langle p_0^2 p_0^2 \rangle \). This parameter takes values between 0 and 1 for states ranging from chaotic/thermal to coherent. Using this parameter the full statistical properties of the ground state are unravelled. Starting with an incoherent state (\( N \) polaritons randomly distributed in the excited level), the coherence rises as the ground state becomes populated. This is clearly shown in Fig. 4a. Again, the most interesting observation about this evolution is the difference between the smooth growing of coherence in the non-interacting regime as compared with the sudden rise of coherence for the interacting case, complementing the results depicted in Fig. 3. A threshold behavior is clearly observed in Fig. 4b. Once the number of polaritons, or equivalently the density, is increased beyond a certain value the coherence rises suddenly. A possible phase transition triggered by Coulomb interactions is thus reinforced. Moreover, since no high order correlations are in principle discarded by the present method, as it is usually the case for condensed matter systems [16,17] (excitons) and [6] (polaritons), we can quantify their influence on the polariton condensation dynamics.

Finally, we would like to remark some of the most important aspects of these results. First of all, the threshold behavior that governs the dynamics was already discussed in Ref.[4], and reported results are in good agreement with ours. This gives us confidence that the stochastic scheme we used is indeed a very practical tool for the study of complex systems like those found in semiconductor microcavities. In particular, effects due to the semiconductor environment are responsible for the relaxation of the system, leading to a continuous accumulation of polaritons in the same final state at low temperatures. Nevertheless, correlations generated by interactions play a crucial role in the polariton condensation dynamics. While a coherence degree parameter close to 1 can only be interpreted as a criteria for polariton lasing [4], instead of BEC in a quasi-two-dimensional nanostructure, signs of a true phase transition are present in our results as a consequence not only of the bosonic behavior but from quantum fluctuations provided by polariton interactions. Since the appearance of the phase transition is governed by Coulomb interactions, our results point toward a BEC type behavior rather than a polariton laser.

4. Polariton parametric scattering

In recent experiments [2,3] polaritons are optically excited at a well defined in-plane momentum \( k \), allowing a direct control of their dynamics. The strong Coulomb in-
interaction results in a spontaneous parametric scattering of polariton pairs (signal and idler) according to momentum and energy conservation criteria. For resonant optical excitation, the scattering leads into a final state $8$-shaped distribution in $\mathbf{k}$ space, as first suggested by Ciuti et al [13] and experimentally confirmed in Ref.[2]. We use the present stochastic scheme to study the dynamics of initially pumped polaritons in a particular momentum state $\mathbf{k}$ in the lower polariton branch. Once again, the advantages of the present approach are manifold. Similar to the condensation of polaritons studied in the previous Section, the theoretical discussion of parametric scattering has been based upon MFA, and thus the results are only valid below the threshold of parametric luminescence. Thus, the study of the macroscopic behavior of interacting polaritons in a relaxing environment has been poorly investigated [9]. Moreover, the dynamical behavior of condensed matter quasi-particles in an out of equilibrium regime can be systematically studied within our approach. In this Section we abandon the effective two-level approximation and resort to an extended multi-level description.

For the numerical simulations that follows, the total Hamiltonian is still given by Eq.6, where the system’s parameters are the same as in Section 3. The possible final states for the scattering of two polaritons pumped at $\mathbf{k} = \{k_p, 0\}$, into a signal and idler pair, are displayed in Fig.5, where the long-time limit, in which both energy and lineal momentum have to be conserved, is depicted as the continuous line. This information is crucial for understanding the many-polariton case that we discuss from now on. The initial state is taken as $N$ polaritons pumped at $\mathbf{k}_p = \{0.66, 0\} \text{µm}^{-1}$. The system is let to evolve and population in $\mathbf{k}$ space is monitored at different times. In Fig.6 contour plots of the polariton population distribution are shown, for $N=1000$ particles initially pumped. Clearly, a strong asymmetry in signal and idler emission patterns is seen. In particular, although there is a small population in the idler modes, it is not comparable to the signal one.

Above the parametric threshold a remarkable different behavior is displayed. As the density of polaritons increases a more significant fraction of the population is concentrated in lowest momentum states, and thus an important intensity is registered in the idler state opposed to the signal one, as can be seen in Fig.7. Similar results were reported in Ref.[9]. Furthermore, the finite intensity that rises in the idler state is asynchronous with the evolution of the signal population, in close agreement with experimental results in Ref.[2]. The asymmetry in the shape and time of the formation dynamics of the final state distribution in $\mathbf{k}$ space is a direct result of the different multiple scattering processes affecting signal and idler polaritons. As is well known, for higher momentum states (idler modes), polaritons have a big excitonic component, which implies a stronger coupling with the lattice phonons, and as a consequence a fast decay in idler population. Even though in the time range of the reported dynamics the dominant scattering processes correspond to Coulomb interactions, some evidence of decoherence in the idler states, due to the semiconductor environment (phonons), is visible in Fig.7c and 7d.

In contrast to MFA, the threshold behavior is well captured by our stochastic method. As the polariton density increases, a dramatic change is observed in the signal population. While below the threshold the signal population is weak, a sudden increase close to the threshold is observed. In Fig.8, the signal population is displayed for different times as a function of the number of initially pumped polaritons. Close to $N = 2000$, a clear change of slope is registered for any time. The behavior of the idler mode populations (not shown) is analogous.
5. Conclusions

In the present paper we have presented a stochastic study on the dynamical properties of microcavity polaritons. In particular, we reviewed the dynamical condensation of polaritons emphasizing on the role of Coulomb interactions. Additionally, the threshold behavior of the spontaneous parametric downconversion of polaritons was also discussed. The numerical calculations have been performed by means of stochastic evolution of a reduced state vector in the single particle Hilbert space, separately from the stochastic environment evolution. The resulting stochastic differential equations allows reasonable reduction of the problem which by direct numerical integration of the density matrix would have being cumbersome. For this matter, the stochastic technique turns into a very practical tool for many body problems in condensed matter scenarios. Its validity was tested and compared positively with previous theoretical and experimental results.

In the dynamical condensation of polaritons, we found that while the semiconductor environment induces a slow relaxation process, polariton-polariton interactions trigger the sudden development of a macroscopic coherent state. This differs clearly from the polariton laser picture, and other recent proposals which claim that this behavior is just a consequence of the particle number conservation \[4\]. While a more detailed study on this phenomenon is needed, our results favor the BEC type phase transition of microcavity polaritons.

On the other hand, the dynamical process of parametric emission was studied. There exists an asymmetry in shape and time dependence of the far field emission of signal and idler modes governed by this kind of scattering processes. The threshold behavior is also well captured by our stochastic method. This corresponds to the sudden rise in signal and idler populations. Coherent properties of this parametric emission process may also be studied by the present method. Additionally, a straightforward possibility is to extend the present stochastic method to finite lattice temperature situations.

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