

Linewidth of a polariton laser: Theoretical analysis of self-interaction effects

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Polaritons in semiconductor microcavities can experience a Bose-Einstein condensation experimentally detectable in the spectrum of the emitted light. Scattering with noncondensed particles as well as self-interaction in the condensate provoke phase diffusion limiting the coherence of the polariton condensate. We present a theoretical analysis of self-interaction effects on the lineshape of the emission from a polariton laser. Our calculations, for CdTe microcavities, show that there is an optimum pump at which the linewidth of the emitted light is reduced down to $1 \mu\text{eV}$.

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A Bose-Einstein atomic condensate is a source of coherent matter waves (matter laser), in the same way as a photonic laser is a source of coherent light. Another system capable of undergoing a similar condensation is that of polaritons formed by the strong coupling between quantum well excitons and confined photons in a semiconductor microcavity. Polaritons behave as composite bosons at densities below the saturation density, as confirmed by recent experiments that include the observation of stimulated scattering, and parametric amplification and oscillation.^{1,2} A polariton Bose-Einstein condensate is a matter laser that can be optically pumped, and experimentally detected by the emitted light.³⁻⁵ The growth of semiconductor microcavities with new materials, such as II-VI compounds, or GaN, opens great possibilities, due to the strong stability of the exciton in these systems.^{6,7} In particular, a recent calculation⁸ shows that in high quality CdTe microcavities, huge occupation numbers can be achieved in the polariton ground state at densities well below the saturation density, i.e., at densities at which polaritons can be described as interacting bosons. Under these conditions, the system would be unstable to symmetry breaking, and thus to the formation of a Bose-Einstein condensate (BEC).

In this Rapid Communication, we establish experimental signatures of a polariton laser. We consider a microcavity pumped by a continuous nonresonant laser, so that the polariton-polariton scattering is fast enough to overcome the radiative losses and the system is able to relax to the ground state. At densities larger than a given threshold for BEC, the lowest energy level shows a macroscopic occupancy, and the system becomes a continuous polariton laser. The standard theory of photon lasers,⁹ i.e., noninteracting systems, predicts a very narrow linewidth, inversely proportional to the number of condensed particles, $\Gamma^{\text{NL}} \propto 1/N_0$. However, polaritons interact with each other through some potential V and new physics is involved. Self-interaction in the condensate provokes a process of phase diffusion that is determined by the energy scale VN_0 .¹⁰⁻¹² When VN_0 is comparable to $\hbar\Gamma^{\text{NL}}$, this process would increase the linewidth as the number of condensed bosons increases, a behavior that is the opposite to that of a photon laser. We introduce here a self-consistent framework to include the two effects described above. We show that there is an optimum pumping range to

get an extremely narrow linewidth of the polariton emission. For CdTe microcavities, we find that the linewidth can be reduced down to $1 \mu\text{eV}$.

Figure 1 depicts schematically the lower energy branch of the spectrum of a semiconductor microcavity for the case of zero detuning.¹⁴ Below the minimum bare exciton energy, ϵ_0^x , excitons and photons are strongly coupled. The ground state ($k=0$) lies at the energy $\Omega_P/2$ below ϵ_0^x , where Ω_P is the polariton splitting. At energies above ϵ_0^x , the polariton branch merges abruptly into the bare exciton dispersion, and the exciton-photon coupling can be neglected. We consider the phase space as divided in two regions: the lower energy states, labeled as lower polaritons (LP), and the states above ϵ_0^x , treated as bare excitons. The small density of states of the LP, when compared to the exciton one ($\rho_{\text{LP}} \approx 10^{-4} \rho_x$), drastically reduces the threshold density for the formation of a BEC in the LP part of the spectrum. This allows us to simplify the problem of the polariton dynamics by considering the exciton states as a thermalized reservoir.⁸ On the other hand, microcavity polaritons are two-dimensional (2D) quasiparticles that can undergo a Kosterlitz-Thouless transition in the thermodynamic limit.¹³ Finite size effects lead to a local transition to BEC, so that we quantize the LP levels according to a scale determined by area S of the spot of the pumping laser.

In this work we are not analyzing the properties of the quantum state of the polariton condensate, but we are inter-

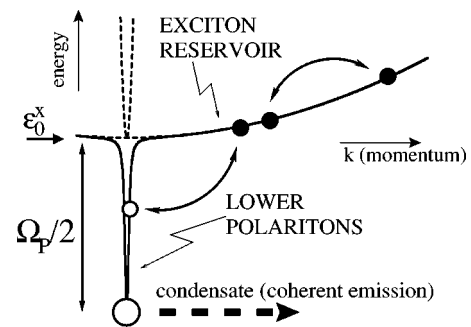


FIG. 1. Phase space for the scattering of polaritons in a semiconductor microcavity. Upper and lower dashed lines represent the bare photon and bare exciton, respectively. The continuous line is the polariton dispersion that results from the strong exciton-photon coupling.

ested in the spectrum of the emitted light, which can be calculated from the adequate correlation function. The steps of the theoretical analysis are: first, to obtain an equation of motion for the density matrix, and later to compute correlation functions and emitted intensities.

Equation of motion for the density matrix. The main mechanism for the relaxation to the lower energy states is the scattering of two excitons,¹⁵ in which one of the final states is a LP. We label this scattering process exciton-polariton (XP) scattering. This relaxation process creates a nonequilibrium polariton distribution that evolves towards a Bose-Einstein condensate as exciton density is increased.⁸ Our Hamiltonian is the sum of three terms describing the bare-exciton and LP dispersions (H_0), the XP scattering (H_{XP}), and the self-interaction in the condensate mode (H_{SI}):

$$H_0 = \sum_k \epsilon_k^{\text{LP}} a_k^\dagger a_k + \sum_k \epsilon_k^x b_k^\dagger b_k,$$

$$H_{XP} = \sum_k a_k^\dagger F_k^\dagger + \text{H.c.}, \quad H_{SI} = V a_0^\dagger a_0^\dagger a_0 a_0. \quad (1)$$

The index k is quantized according to S . a_k^\dagger (ϵ_k^{LP}) and b_k^\dagger (ϵ_k^x) are the creation operators (energies) of the LP and bare excitons, respectively. $F_k^\dagger = \sum_{k_2, k_3, k_4} V_{k, k_2, k_3, k_4} b_{k_2}^\dagger b_{k_3}^\dagger b_{k_4}$ describes the scattered excitons, with V_{k_1, k_2, k_3, k_4} being the polariton-polariton interaction in the $ka_B \ll 1$ limit (see Ref. 16 for an explicit expression). $V = V_{0,0,0,0}$ is the self-interaction in the ground state. We have neglected polariton-polariton scattering that involves more than one lower polariton and polariton-phonon scattering, because they are much slower than the process depicted in Fig. 1, and only produce energy shifts.^{8,16}

Since we are mainly interested in the evolution of LP, we trace out the reservoir degrees of freedom in the density matrix operator χ , and define the reduced density matrix s :

$$s(t) = \text{Tr}_R\{\chi(t)\}, \quad \langle O_{\text{LP}}(t) \rangle = \text{Tr}_{\text{LP}}\{O_{\text{LP}}(t)s(t)\}, \quad (2)$$

where Tr_R and Tr_{LP} represent the trace over the exciton reservoir and the LP, respectively, and O_{LP} is any function of LP operators.

We describe the exciton reservoir by a thermalized Maxwell-Boltzmann distribution. This approximation is justified by the fast exciton-exciton scattering within the exciton reservoir. Moreover, this assumption is supported by a recent experiment¹⁷ on the evolution of the polariton distribution when pump power or temperature is varied. The total density matrix operator can be approximated by

$$\chi(t) \approx s(t) \otimes f_R(t) = s(t) \otimes \exp\left(\sum_k (\mu_x - \epsilon_k^x) b_k^\dagger b_k / k_B T_x\right), \quad (3)$$

where μ_x and T_x are the chemical potential and temperature in the exciton reservoir, respectively.

The time evolution of the density matrix is calculated in the interaction picture. Up to the lowest order in $H_{XP} + H_{SI}$, s evolves as

$$\begin{aligned} \frac{d}{dt}s(t) = & \frac{1}{i\hbar}[H_{SI}, s] - \frac{1}{\hbar^2} \int_{t_0}^t dt' \sum_{k,q} (a_k a_q^\dagger s' - a_q^\dagger s' a_k) \\ & \times \langle F_k F_q^\dagger \rangle + (a_k^\dagger a_q s' - a_q s' a_k^\dagger) \langle F_k^\dagger F_q \rangle \\ & + (s' a_q^\dagger a_k - a_k s' a_q^\dagger) \langle F_q^\dagger F_k \rangle \\ & + (s' a_q a_k^\dagger - a_k^\dagger s' a_q) \langle F_q F_k^\dagger \rangle, \end{aligned} \quad (4)$$

where all operators are in the interaction picture with respect to $H_{XP} + H_{SI}$ and the primes indicate time dependence on t' .

Equation (4) becomes simpler when LP operators at time t are translated to the previous time t' by $a_k^\dagger(t) = a_k^\dagger(t') e^{i\epsilon_k^{\text{LP}}(t-t')/\hbar}$. Moreover, we are interested on the steady-state regime, i.e., in the limit $t_0 \rightarrow -\infty$, so that a Markovian approximation is well justified. Under this approximation, we take the LP operators in Eq. (4) out of the time integration and obtain the following master equation:

$$\begin{aligned} \frac{ds}{dt} = & \frac{iV}{\hbar}[s, a_0^\dagger a_0^2] + \sum_k \left(\frac{W_k^{\text{in}}}{2} (a_k^\dagger s a_k - a_k a_k^\dagger s) \right. \\ & \left. + \frac{W_k^{\text{out}} + \Gamma_k}{2} (a_k s a_k^\dagger - a_k^\dagger a_k s) + \text{H.c.} \right). \end{aligned} \quad (5)$$

The rates W_k^{in} and W_k^{out} are easily evaluated with the thermalized exciton distribution:⁸

$$\begin{aligned} W_k^{\text{in}} = & \sum_{k_2, k_3, k_4} 4 |V_{k, k_2, k_3, k_4}|^2 (1 + N_{k_2}^x) N_{k_3}^x N_{k_4}^x \\ & \times \delta(\epsilon_k^{\text{LP}} + \epsilon_{k_2}^x - \epsilon_{k_3}^x - \epsilon_{k_4}^x), \end{aligned} \quad (6)$$

$$\begin{aligned} W_k^{\text{out}} = & \sum_{k_2, k_3, k_4} 4 |V_{k, k_2, k_3, k_4}|^2 N_{k_2}^x (1 + N_{k_3}^x) (1 + N_{k_4}^x) \\ & \times \delta(\epsilon_k^{\text{LP}} + \epsilon_{k_2}^x - \epsilon_{k_3}^x - \epsilon_{k_4}^x), \end{aligned}$$

where N_k^x are the occupancies in the exciton reservoir. The imaginary parts of the time integrations have been neglected because they lead to energy shifts irrelevant for the process of BEC and phase diffusion that we consider here. In Eq. (5), we have also included a term [not appearing⁹ in Eq. (4)] that accounts for the radiative losses with a rate $\Gamma_k = C_k^{\text{LP}}/\tau_{\text{ph}}$, where C_k^{LP} is the photon weight in the polariton wave function and τ_{ph} is the lifetime of the photonic mode confined in the microcavity.

Equation (5) is the keynote of our analysis. In particular, it allows to calculate the evolution (which does not depend on the self-interaction) of the LP occupation numbers:

$$\frac{d}{dt} \langle a_k^\dagger a_k \rangle = W_k^{\text{in}} (\langle a_k^\dagger a_k \rangle + 1) - (W_k^{\text{out}} + \Gamma_k) \langle a_k^\dagger a_k \rangle. \quad (7)$$

We also compute self-consistently the evolution of the parameters n_x , T_x that describe the exciton reservoir, by means of the corresponding rate equations as described in Ref. 8. From the steady n_x , T_x , we get the rates W_0^{in} , W_0^{out} that

correspond to the scattering to the ground state. After that, we have found all the parameters in the master equation (5).

Emission spectrum. Once we have a self-consistent description for the time evolution of the density matrix, we can undertake the task of computing expectation values of magnitudes experimentally measurable. In particular the emission spectrum can be obtained from the two-time correlation function:

$$I(\epsilon) = \frac{1}{\pi} \text{Re} \int_0^\infty \langle a_0^\dagger(\tau) a_0(0) \rangle e^{-i\epsilon\tau/\hbar} d\tau. \quad (8)$$

Without self-interaction in Eq. (5), the application of the quantum regression theorem would lead trivially to Lorentzian line shape of the spectrum:

$$\langle a_0^\dagger(\tau) a_0(0) \rangle^{\text{NI}} = N_0 e^{-\Gamma^{\text{NI}}\tau}, \quad \Gamma^{\text{NI}} = \frac{W_0^{\text{out}} + \Gamma_0}{2(1 + N_0)}. \quad (9)$$

Decoherence has the usual aspect of a single-particle noise corrected, in the denominator, by the number of particles in the condensate. However, inclusion of the self-interaction term changes this result dramatically. We use a well-known method in quantum optics that allows us to include exactly the effect of the self-interaction.⁹ The two-time average is expanded as a sum over a set of auxiliary functions $C_n(\tau)$:

$$\langle a_0^\dagger(\tau) a_0(0) \rangle = \sum_n \sqrt{n} C_n(\tau),$$

$$C_n(\tau) = \langle e^{iH_0\tau/\hbar} |n\rangle \langle n-1 | e^{-iH_0\tau/\hbar} a_0(0) \rangle, \quad (10)$$

with $|n\rangle$ being the number representation of the $k=0$ mode. One can use the quantum regression theorem to show that the functions C_n satisfy the differential equation¹⁰

$$\frac{d}{d\tau} C_n(\tau) = \frac{i}{\hbar} \epsilon_0^{\text{LP}} C_n(\tau) + \sum_m L_{n,m} C_m(\tau), \quad (11)$$

where the $L_{n,m}$ are the coefficients governing the evolution of off-diagonal one-time matrix elements:

$$\frac{d}{d\tau} \langle n-1 | s(\tau) | n \rangle = \sum_m L_{n,m} \langle m-1 | s(\tau) | m \rangle. \quad (12)$$

Using Eqs. (5) and (12), Eq. (11) becomes

$$\begin{aligned} \frac{d}{d\tau} C_n(\tau) = & \left(-w_0^+(2n+1) - w_0^-(2n-1) + \frac{i}{\hbar} V(n-1 \right. \\ & \left. - N_0) \right) C_n(\tau) + w_0^+ 2\sqrt{n(n-1)} C_{n-1}(\tau) \\ & + w_0^- 2\sqrt{n(n+1)} C_{n+1}(\tau), \end{aligned} \quad (13)$$

with $w_0^+ = W_0^{\text{in}}/2$ and $w_0^- = (W_0^{\text{out}} + \Gamma_0)/2$. In Eq. (13) we have taken the origin of energies at $\epsilon_0^{\text{LP}} + VN_0$, in order to compare the linewidths of the emission spectrum at different densities. Initial condition for Eq. (13) is obtained from C_n for $\tau=0$:

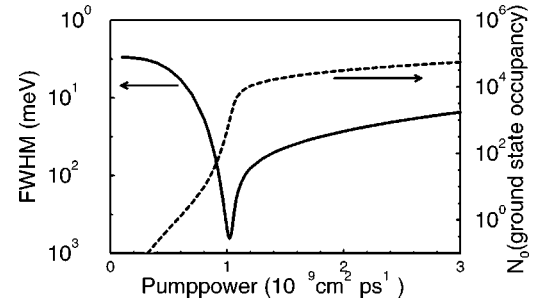


FIG. 2. Full width at half maximum of the emission spectra (continuous line) and ground state occupancy (dashed line) as a function of the pump power p_x (in $10^9 \text{ cm}^{-2} \text{ ps}^{-1}$). The optimum pump corresponds to $p_x \approx 10^9 \text{ cm}^{-2} \text{ ps}^{-1}$.

$$C_n(0) = \sqrt{n} \langle n | s | n \rangle = \sqrt{n} \left(1 - \frac{w_0^+}{w_0^-} \right) \left(\frac{w_0^+}{w_0^-} \right)^n, \quad (14)$$

where $\langle n | s | n \rangle$ is easily deduced from having a time derivative equal to zero in the steady state. Instead of solving numerically the enormous set of Eqs. (13), the problem can be simplified, in the case $N_0 \gg 1$, by replacing the index n by a continuous variable,¹⁸ obtaining a partial differential equation:

$$\begin{aligned} \frac{\partial C(n, \tau)}{\partial \tau} = & n(w_0^- + w_0^+) \frac{\partial^2}{\partial n^2} C(n, \tau) + [2n(w_0^- - w_0^+) + w_0^- \\ & + w_0^+] \frac{\partial}{\partial n} C(n, \tau) + \left(2(w_0^- - w_0^+) - \frac{w_0^- + w_0^+}{4n} \right. \\ & \left. + \frac{iV}{\hbar} (n - N_0 - 1) \right) C(n, \tau). \end{aligned} \quad (15)$$

In the limit $\hbar\Gamma^{\text{NI}} \gg VN_0$, Eq. (15) gives the adequate Lorentzian shape of the spectrum with a linewidth Γ^{NI} . In the opposite limit, $VN_0 \gg \hbar\Gamma^{\text{NI}}$, an analytic solution exists:

$$C(n, \tau) \approx \frac{1}{N_0} \sqrt{n} e^{-n/N_0 + i(V/\hbar)(n - N_0)\tau}, \quad (16)$$

predicting an asymmetrical line shape:

$$I(\epsilon) \approx \frac{\hbar}{V} \frac{(\epsilon + VN_0)}{VN_0} e^{-(\epsilon + VN_0)/(VN_0)} \theta(\epsilon + VN_0). \quad (17)$$

Results for a CdTe microcavity. We have solved numerically Eq. (15) for the case of a CdTe microcavity with $\Omega_p = 10 \text{ meV}$, and zero detuning between the bare exciton and the photonic mode. The pump is assumed to add excitons at a given rate, p_x , and at a lattice temperature, $T_L = 10 \text{ K}$. This implies a very fast relaxation by the exciton-phonon scattering, which is not always the case in experiments. The steady value of T_x , however, can reach 30 K, as explained in Ref. 8. Lifetimes of the photon and the bare excitons are taken $\tau_{\text{ph}} = 1 \text{ ps}$ and $\tau_x = 100 \text{ ps}$. The steady-state polariton density considered in our calculation is always below 0.3 times the saturation density, which can be estimated to be $6.7 \times 10^{11} \text{ cm}^{-2}$ in a CdTe microcavity.⁸ The quantization

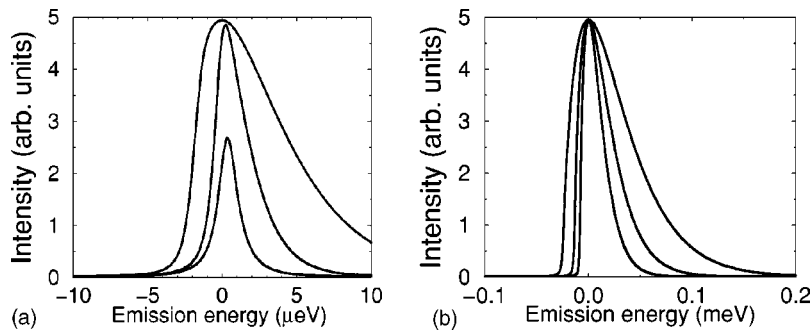


FIG. 3. (a) Transition from noninteracting to self-interaction decoherence in the emission spectra for $p_x=1.02$ (inner), 1.05, 1.10 (outer) in units of $10^9 \text{ cm}^{-2} \text{ ps}^{-1}$. (b) Continuous increase of the linewidth shown in the spectra for $p_x=1.5$ (inner), 2.0, 3.0 (outer) in units of $10^9 \text{ cm}^{-2} \text{ ps}^{-1}$.

length is $50 \mu\text{m}$, of the order of typical excitation spot diameters. Figure 2 gives N_0 as a function of the pump power. It shows a threshold for BEC ($N_0 > 1$) around $p_x \approx 810^8 \text{ cm}^{-2} \text{ ps}^{-1}$.

For densities below the condition $N_0 V \approx \hbar \Gamma^{\text{NI}}$, the calculated line shape is Lorentzian with minimum linewidth of the order of $1 \mu\text{eV}$. After condition $N_0 V \approx \hbar \Gamma^{\text{NI}}$ is reached, there is a transition from the laserlike decoherence to the self-interaction broadening as shown in Fig. 3(a). For larger pump powers, the emission is asymmetric and is continuously broadened. Above $p_x = 1.110^9 \text{ cm}^{-2} \text{ ps}^{-1}$, the numerical results are identical to the approximation given in Eq. (17). In the evolution of the spectrum linewidth as a function of the pump power, shown in Fig. 2, one observes that the optimum pump is very well defined by the abrupt dip in the emission linewidth. This dip means an increase of the coherence of almost three orders of magnitude. For larger densities the linewidth of the polariton laser increases linearly with the

number of condensed particles, until it reaches values comparable to the noncondensed emission.

In conclusion, we have presented a theoretical method for the self-consistent calculation of the emission linewidth of a polariton BEC. Self-interaction sets an important restriction for the coherence that can be achieved in this system. The dynamics described by our Eq. (5) would drive the system to a BEC provided a small coherent seed, i.e., a symmetry-breaking term, is included in the initial condition. Our conclusions are also relevant for the case of recent proposals in which the scattering mechanism for the relaxation of polaritons towards the ground state is different than the XP scattering considered here, as for instance the case of the proposed electron-polariton scattering in doped microcavities.¹⁹

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