Dynamical condensation of polaritons

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We develop a few-level quantum theory of microcavity polaritons in presence of both Coulomb and polariton-phonon interaction, obeying particle number conservation. We study the growth of a macroscopic population of condensed particles in the lowest polariton state, under steady-state incoherent excitation of higher energy states. The Coulomb interaction drives the quantum fluctuations that finally trigger the transition to condensed phase, while the polariton-phonon interaction results in a Boltzmann dynamics that induces a lower state occupation. The transition occurs below the optical saturation density, as in recent experimental findings.

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Quantum fluids are the most remarkable manifestation of quantum mechanics at the macroscopic scale. Superconductivity, superfluidity [1] and more recently Bose-Einstein condensation (BEC) of diluted atoms [2] are all examples of a system in which many particles share the same quantum mechanical wave function. A long sought and never observed quantum fluid is the BEC of excitons in semiconductors [3, 4]. It is believed that exciton BEC is hindered by the very efficient scattering mechanisms – both elastic on defects and impurities and inelastic on e.g. phonons – which induce localization and decoherence. On the other hand, the possibility of achieving a quantum fluid in a solid-state device, with ease of control and integration, would open a new promising way to the implementation of quantum information technology [5].

Recently, it was suggested that a quantum phase transition of polaritons in a semiconductor microcavity under steady-state incoherent optical pumping might occur, with formation of a collective state of many polaritons [6, 7, 8, 9, 10]. The interest of this system resides in the mixed nature of polaritons, which are a linear superposition of photon and exciton states. The very light polariton effective mass implies a long polariton coherence length and robustness to scattering processes. The exciton content, on the other hand, is responsible for polariton-polariton interaction. This nonlinearity is eventually expected to trigger the quantum phase transition.

A parallel between this mechanism and conventional BEC is made hazardous by the Mermin-Wagner theorem [11], stating that a phase transition with an evident symmetry breaking is forbidden in a 2-dimensional system. For this reason, the phenomenon has been rather interpreted as a *polariton laser* transition [9, 10]. A few experimental results suggest the occurrence of this transition [6, 7, 8], but the unexpected observation of a thermal-type intensity correlation function far above threshold [8], doesn't match the laser picture [12]. Laussy et al. [10] have pointed out that an important role is played by the particle number conservation. Indeed, in any symmetry breaking approach, a state with a well defined quantum phase cannot be stationary, due to the fluctuations of the particle number [13]. Therefore, as in the theory of BEC in diluted atoms, a number-conserving approach is needed in order to correctly describe the quantum phase diffusion of the condensate [13, 14, 15]. In order to investigate the appearance of condensation (either at finite temperature or in a non-equilibrium regime), it is important to remark that in the BEC models [1, 2, 16] both the condensed and the non-condensed phases, having different fluctuation terms, are considered. In these models, the interactions are the key feature inducing the phase transition (leading to the condensate) [16]. On the theoretical side, the existing works prefer overlooking this aspect [9, 10], pursuing a strict analogy with the laser theory.

In this letter we develop a few-level model of the polariton dynamics which includes the polariton-polariton Coulomb interaction and the polariton-phonon scattering on equal grounds, considering a non-equilibrium steady-state optical pump populating the high energy state. The model is solved within the Hartree-Fock-Bogoliubov (HFB) approximation, as in the case of quantum fluids at finite temperature [17, 18], but imposing the particle number conservation [14, 15]. The resulting polariton dynamics, under steady-state excitation of an incoherent polariton population, displays a transition at a finite intensity. At threshold the population of the condensed phase increases dramatically. Above threshold, while the population of the non- condensed phase displays an upper bound, the condensate grows linearly. We show that the polariton-polariton interaction is responsible for this peculiar behavior.

The Hamiltonian for the lower polariton in presence of Coulomb and polariton-phonon scattering is [19, 20]

$$H = \sum_k \omega_k \hat{p}_k^\dagger \hat{p}_k + \sum_q \omega_q b_q^\dagger b_q + H_C + H_{ph} \quad (1)$$

$$H_C = \frac{1}{4} \sum_{kk'q} v_{kk'}^{(q)} \hat{p}_{k+q}^\dagger \hat{p}_{k'-q}^\dagger \hat{p}_{k'} \hat{p}_k \quad (2)$$

$$H_{ph} = \sum_{kk'q} g_{kk'}^{(q)}(b_q^\dagger + b_{-q})(\hat{p}_k^\dagger \hat{p}_{k'} + \hat{p}_{k'}^\dagger \hat{p}_k)\,, \qquad (3)$$

where $\hat{p}_k$ defines a Bose field $[\hat{p}_k, \hat{p}_{k'}^\dagger] = \delta_{kk'}$. The polariton operator $\hat{p}_k = P_k \hat{a}_k + \tilde{p}_k$ is expressed as the sum of a condensate part $P_k \hat{a}_k$ and a fluctuation term $\tilde{p}_k$ [15]. The condensed particle operator obeys the Bose commutation relations $[\hat{a}_k, \hat{a}_k^\dagger] = 1$ and $n_c(k) = \langle \hat{a}_k^\dagger \hat{a}_k \rangle$ defines the number of condensed particles in the state with momentum k, while the amplitude P_k represents the wave function of the condensate in the configuration space. In a standard quantum-field approach [15], this wave function is determined self-consistently by the Gross-Pitaevskii equation. The fluctuation part $\tilde{p}_k$ obeys the commutation relation $[\tilde{p}_k, \tilde{p}_k^\dagger] = 1- \mid P_k \mid^2$. The operators are treated in the Heisenberg representation. The total population is $N_{kk} = \langle \hat{p}_k^\dagger \hat{p}_k \rangle = \mid P_k \mid^2 n_c(k) + \tilde{N}_{kk}$, where $\tilde{N}_{kk} = \langle \tilde{p}_k^\dagger \tilde{p}_k \rangle$ is the non-condensed population. Condensed particles are assumed in the lowest state only, $n_c(k) = n_c \delta_{0,k}$, while we assume $\hat{p}_k = \tilde{p}_k$ for $k \neq 0$. In the number-conserving HFB treatment, only processes creating (or annihilating) pairs of condensed particles are taken into account. In this approximation the Coulomb Hamiltonian is written as [17]

$$\begin{aligned} H_C &= \frac{v}{4}\sum_k \{2(N_{00} + \sum_{k'\neq 0} \tilde{N}_{k'k'})\hat{p}_k^\dagger \hat{p}_k + \\ &+ [(P_0^*)^2 \hat{a}_0^\dagger \hat{a}_0^\dagger + (1-\delta_{k,0})\tilde{p}_0^\dagger \tilde{p}_0^\dagger]\tilde{p}_k \tilde{p}_k\} + h.c. \quad (4)\end{aligned}$$

The first line contains the mean field population terms according to the Hartree-Fock approximation, while in the second line no mean-field approximation has been introduced, in order to allow for particle number conservation. In order to conserve momentum, the only relevant amplitudes are $\tilde{m}_k = \langle \hat{a}_0^\dagger \hat{a}_0^\dagger \tilde{p}_k \tilde{p}_k \rangle$ and $\tilde{m}_{0k} = \langle \tilde{p}_0^\dagger \tilde{p}_0^\dagger \tilde{p}_k \tilde{p}_k \rangle$. These terms correspond to Coulomb scattering diagrams in the theory of interacting bosons [16]. The time evolution of the number of condensed particles n_c, of the populations $\tilde{N}_{kk}(t)$ and of the scattering amplitudes $\tilde{m}_k$ and $\tilde{m}_{0k}$, can be evaluated by means of the Heisenberg equations of motion. In the following we denote $b_q^{(1)} = b_q^\dagger$,$b_q^{(2)} = b_q$, $\hat{p}_k^{(1)} = \hat{p}_k^\dagger$ and $\hat{p}_k^{(2)} = \hat{p}_k$). Consistently with the HFB Hamiltonian, we factor all the higher order correlations according to the self consistent mean field approximation [18]. For example $\hat{a}_0^\dagger \hat{a}_0^\dagger \hat{p}_k^\dagger \hat{p}_k \tilde{p}_0 \tilde{p}_0 = N_{kk}\tilde{m}_0$.

Due to polariton-phonon interaction, phonon-assisted correlations are coupled to the HFB variables. The equations for the first order phonon-assisted correlations $\langle \hat{a}_0^\dagger b_q^{(i)} \hat{p}_k^{(j)} \rangle$ and $\langle b_q^{(i)} \hat{p}_k^{(j)} \hat{p}_{k'}^{(l)} \rangle$ $(i, j, l = 1, 2)$ are formally solved within the self-consistent Markov approximation and the result is plugged into the HFB equations. In this way, the polariton-phonon coupling introduces effective phonon-mediated polariton-polariton interaction terms, the lowest-order ones being proportional to $|g_{kk'}^{(q)}|^2$. The higher order phonon assisted correlations are factored according to the mean field approximation. For example $\hat{a}_0^\dagger \hat{a}_0^\dagger b_q^\dagger b_{-q'} \tilde{p}_0 \tilde{p}_0 = \delta_{q,-q'} n_q \tilde{m}_0$. The phonon populations n_q are assumed to be thermally distributed at the lattice temperature.

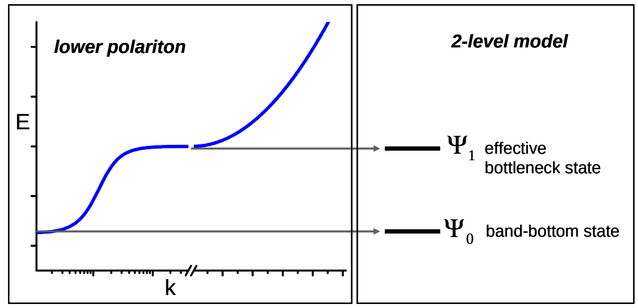

FIG. 1: Lower polariton dispersion. The relaxation between the bottleneck region and the band bottom is described by a 2-level model in which the bottleneck state has an effective population given by ρ_X.

These prescriptions give a closed set of coupled equations for n_c, $\tilde{N}_{kk}$, $\tilde{m}_k$, $\tilde{m}_{0k}$. The solution of the whole set of equations is a cumbersome task. In a typical photoluminescence experiment under nonresonant excitation, the steep polariton dispersion results in a relaxation bottleneck [21, 22], with polariton population piling up at the boundary of the flat exciton-like region of the polariton dispersion. From there, polariton which relax to the band-bottom immediately recombine by emitting a photon. At moderate densities [19], the multiple scattering between polaritons can be neglected. Under these conditions, it is reasonable to assume that the bottleneck states and the state at $k = 0$ obey a closed dynamics. We therefore restrict to a simplified two-level model, sketched in Fig. 1, which describes the coupling between the polariton state at the band bottom and one effective higher energy state in the bottleneck region. The creation operator for the band bottom state is $\hat{p}_0^\dagger$ whereas that for the bottleneck state is $\hat{p}_1^\dagger$. As in previous treatments [23], the high energy state is considered as an effective state spanning the whole bottleneck region. For this reason, all the matrix elements involving this state are renormalized by the total population ρ_x in the bottleneck region. This latter is estimated from the assumption of a thermalized polariton distribution at the bottleneck [23], and is related to the quantization area $A = L_c^2$, the exciton mass M_{exc} and the exciton energy thermal broadening $E \approx k_B T$ by

$$\rho_X = (A/2\pi)(M_{exc}E/\hbar^2), \qquad (5)$$

resulting in an effective phonon coupling strength G [22]. We assume intrinsic linewidths γ_0 and γ_1 for the two levels, accounting for radiative recombination as well as nonradiative homogeneous and inhomogeneous energy broadening. The closed set of six equations for the HFB variables are

$$
\begin{aligned}
\dot{n}_c &= -2\left|P_0\right|^2\left[\gamma_0+G\Gamma_c(n_q-N_{11})\right]n_c+2G\left|P_0\right|^2\Gamma_c(1+n_q)N_{11}+v\mathrm{Im}[(P_0^*)^2(\tilde{m}_0+\tilde{m}_1)] \\
\dot{\tilde{N}}_{00} &= -2Q[\gamma_0+G\tilde{\Gamma}_0(n_q-N_{11})]\tilde{N}_{00}+2GQ^2\tilde{\Gamma}_0(1+n_q)N_{11}+vQ\mathrm{Im}[\tilde{m}_{01}-(P_0^*)^2\tilde{m}_0] \\
\dot{\tilde{N}}_{11} &= -2[\gamma_1+G\Gamma_1(1+n_q+N_{00})]\tilde{N}_{11}+2G\Gamma_1 n_q N_{00}-v\mathrm{Im}[\tilde{m}_{01}+(P_0^*)^2\tilde{m}_1]+F \\
\dot{\tilde{m}}_0 &= -2\{\gamma_0+G(\left|P_0\right|^2\zeta_0+Q\tilde{\zeta}_0)(n_q-N_{11})+i(2Q-1)[\omega_0+v(N_{00}+N_{11})]\}\tilde{m}_0+ \\
&+\ iv[P_0^2 n_c(\tilde{m}_{01}^*+2\tilde{N}_{00}^2)-Q\tilde{N}_{00}(\tilde{m}_1+2P_0^2n_c^2)] \\
\dot{\tilde{m}}_1 &= -2\{\left|P_0\right|^2\left[\gamma_0+G\zeta_1(n_q-3N_{11})\right]+\gamma_1+G\tilde{\zeta}_1(1+n_q+N_{00})+i[\omega_1-|P_0|^2\omega_0+vQ(N_{00}+N_{11})]\}\tilde{m}_1+ \\
&+\ iv[P_0^2n_c(\tilde{m}_{01}+2\tilde{N}_{11}^2)-\tilde{N}_{11}(\tilde{m}_0+2P_0^2n_c^2)] \\
\dot{\tilde{m}}_{01} &= -2\{Q[\gamma_0+G\tilde{\zeta}_{01}(n_q-3N_{11})]+\gamma_1+G\zeta_{01}(1+n_q+3N_{00}-2n_c)+i[\omega_1-Q\omega_0+v|P_0|^2(N_{00}+N_{11})]\}\tilde{m}_{01}+ \\
&+\ iv[Q\tilde{N}_{00}(\tilde{m}_1+2\tilde{N}_{11}^2)-\tilde{N}_{11}(P_0^2\tilde{m}_0^*+2\tilde{N}_{00}^2)].
\end{aligned}
\tag{6}
$$

where $\mid P_0 \mid^2= n_c/(n_c+\tilde{N}_{00})$ and we have indicated $Q=1-|P_0|^2$, $\Gamma_1=\gamma_1/(\gamma_1+\gamma_0)$, $\Gamma_c=(\gamma_1/(\gamma_1+|P_0|^2\gamma_0)$, $\zeta_0=\gamma_1/[\gamma_1+(1+Q)\gamma_0]$, $\zeta_1=\gamma_1/(3\gamma_1+|P_0|^2\gamma_0)$, and $\tilde{\zeta}_1=\gamma_1/[\gamma_1+(3-2Q)\gamma_0]$ (the remaining terms $\tilde{\Gamma}_0$, $\tilde{\zeta}_0$, $\tilde{\zeta}_{01}$, ζ_{01} are obtained exchanging $|P_0|^2$ and Q in Γ_c, ζ_0, ζ_1, $\tilde{\zeta}_1$, respectively). We have denoted by n_{01} the phonon population at the energy matching the gap between the two levels. $|P_0|^2$ is related self-consistently to the condensate fraction in according to [15] as previously noted.

These equations show some interesting properties. The coupling between the condensate population and the correlations, responsible of the instability that drives the condensate buildup, is mediated by the Coulomb interaction, as expected. The phonon-mediated interaction induces a renormalization of the lifetimes and Boltzmann relaxation terms [22]. For the numerical evaluation, parameters of a typical AlAs/GaAs microcavity have been used, with a quantization size of $A=100\ \mu\mathrm{m}^2$. This parameter enters the definition of the Coulomb [20] and phonon [22] coupling terms as well as the expression for ρ_x [23, 24]. A lattice temperature $T=10$ K is assumed. The bare polariton linewidths are taken to be $\gamma_0=0.2$ meV, $\gamma_1=1$ meV. Assuming a Rabi splitting $\Omega_R=3.5$ meV, the Coulomb matrix element is evaluated to be $v=5\times10^{-4}$ meV, while the resulting effective phonon coupling strength (taking into account the bottleneck population) is $G=10^{-2}$ meV. For an energy gap $\omega_1-\omega_0=2$ meV (negative cavity-exciton detuning), we estimate a bottleneck population $\rho_x=10^4$ [23] and a phonon population $n_{01}=0.1$. We introduce a constant F, representing a steady-state source of polariton population produced at the bottleneck after relaxation from higher-energy states, as in a typical experiment with non-resonant CW excitation [6, 7, 8]. We use a small seed for the amplitude P_0 in order to trigger the non trivial solution with $n_c\neq0$. We have carefully checked that the results are independent of the value chosen for this seed.

We have solved numerically the set of equations as a function of time. For each value of the pump amplitude, we observe after a transient a stationary solution for all the quantities. In the following, we discuss these stationary values as a function of the pump intensity F. Fig. 2(a) displays the stationary populations as a function of the normalized pump intensity. A threshold occurs at $F=F_{th}$, for which $n_c\simeq1$. At threshold, the population of bottleneck polaritons $\tilde{N}_{11}$ saturates at a value of 20 polaritons per mode, i.e. to an exciton density of $2\times10^{11}\,\mathrm{cm}^{-2}$, close to the exciton optical saturation density. However, the experimental evidence was obtained in a sample containing 12 quantum wells [8]. In such a situation, the predicted threshold density per quantum well is $\sim10^{10}\,\mathrm{cm}^{-2}$, significantly lower than saturation and in fairly good agreement with the experimental estimate [8]. Above threshold, the condensate population n_c grows linearly whereas the non condensed polariton population $\tilde{N}_{00}$ decreases asymptotically to a finite value. Hence, for pump intensities well above threshold, the condensate fraction approaches unity, as also indicated by the behaviour of $|P_0|^2$ plotted in Fig. 2(b). Our finding that the condensate fraction approaches unity only at $F\gg F_{th}$ might provide an interpretation of the unexpectedly slow decrease of the second-order coherence observed as a function of pump intensity [8]. An evaluation of the second order correlation function in the framework of the present theory represents a cumbersome task and is currently in progress. Fig. 2(c) displays the steady-state quantum correlations $\tilde{m}_0$, $\tilde{m}_1$, and $\tilde{m}_{01}$. These quantities are negligible below threshold but dramatically increase at threshold, thus triggering the phase transition, as expected according to both laser [12] and BEC [1, 2, 3] quantum theories. In order to clarify the role of the Coulomb interaction, we compare in Fig. 2(a) and (b) the steady state solutions obtained neglecting all Coulomb terms in (6). By inspection of Eqs. (6) it is clear that the populations obey a standard Boltzmann dynamics, while the correlations $\tilde{m}_0$, $\tilde{m}_1$, and $\tilde{m}_{01}$ admit a vanishing solution. Without Coulomb interaction, therefore, a standard three-level Boltzmann equation is

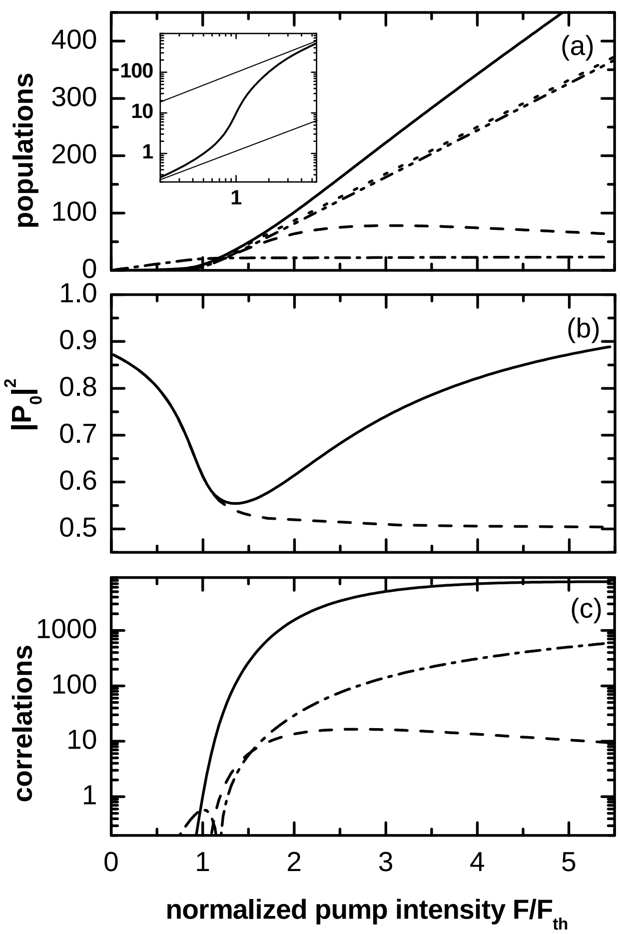

FIG. 2: The steady state solutions plotted as functions of the normalized pump intensity. (a) Solid line: the condensate population n_c (plotted also on a log-log scale in the inset). Dashed line: the non condensed low-energy population $\tilde{N}_{00}$. Dot-dashed line: the high-energy population $\tilde{N}_{11}$. The same quantities n_c (dotted) and $\tilde{N}_{00}$ (dot-dot-dashed), computed neglecting the Coulomb interaction, are also plotted. (b) The condensate fraction $|P_0|^2$ as obtained from the full solution (solid line) and neglecting the Coulomb interaction (dashed line). (c) The Coulomb correlations $|\tilde{m}_0|$ (solid line), $|\tilde{m}_1|$ (dashed line) and $|\tilde{m}_{01}|$ (dot-dashed line).

recovered. As a consequence, the equations for n_c and $\tilde{N}_{00}$ have identical form and result in a similar dynamics for the two quantities. This is shown in Fig. 2 (a) where both n_c and $\tilde{N}_{00}$ grow linearly above threshold, and in Fig. 2 (b) where $|P_0|$ reaches a stationary value of only 0.5. From this comparison the important role played by the Coulomb interaction in determining the phase transition behaviour of the system is clear.

In conclusion, within a few-level model we have described the dynamical condensation of microcavity polaritons. Our theory is analogous to the quantum field description of BEC of a diluted atom gas, accounting for the coexistence of a condensed and a non-condensed phase. The phase transition at threshold is triggered by quantum fluctuations via the Coulomb interaction. In this respect, our result is more closely related to BEC than the polariton laser theory [9, 10], in which the polariton field is treated according to the quantum theory of a single-mode laser. Like both number conserving BEC [15] and laser [12] theories, our treatment predicts zero expectation value of the condensate quantum field $\langle \hat{a}_0^\dagger \rangle$, and thus preserves the $U(1)$ phase symmetry. The condensate wave function P_0 is nonvanishing even below threshold, due to fluctuations, and represents therefore an order parameter only in a weak sense, in analogy with the average electric field amplitude in the quantum laser theory [12]. On this ground, the mechanism we describe might be considered as a polariton laser. However, the coexistence of the two phases, as well as the Coulomb interaction play a crucial role in the condensation dynamics and allow a satisfactory account of experimental results.

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