Excitations in a non-equilibrium Bose-Einstein condensate of exciton-polaritons

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We have developed a mean-field model to describe the dynamics of a non-equilibrium Bose-Einstein condensate of exciton-polaritons in a semiconductor microcavity. The spectrum of elementary excitations around the stationary state is analytically studied in different geometries. A diffusive behaviour of the Goldstone mode is found in the spatially homogeneous case and new features are predicted for the Josephson effect in a two-well geometry.

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After a few decades of impressive efforts on a variety of different systems such as bulk cuprous oxide \cite{1} and coupled quantum wells \cite{2}, first observations of Bose-Einstein condensation (BEC) of excitons in solid state systems have been recently reported in a gas of exciton-polaritons \cite{3} and immediately confirmed by other groups \cite{4, 5}. The system under investigation consists of a semiconductor microcavity containing a few quantum wells with an excitonic transition strongly coupled to the cavity photon mode. In this strong coupling regime, the basic excitations of the system are exciton-polaritons, i.e. linear superpositions of a quantum well exciton and a cavity photon. As compared to other examples of BEC, namely in liquid $^4$He and ultracold atomic gases, the main novelty of the present polariton system is its intrinsic non-equilibrium nature due to the finite lifetime of polaritons. The condensate has in fact to be continuously replenished from the relaxation of optically injected high energy excitations (e.g. free carriers or hot polaritons), and its steady state results from a dynamical equilibrium between pumping and losses. This makes the present system a unique candidate for the study of the BEC phase transition in a non-equilibrium context. Recent theoretical work \cite{6} has suggested that the non-equilibrium condition is responsible for dramatic changes in the dispersion of low-lying excitations of incoherently pumped polariton condensates: the sound mode of equilibrium condensates is replaced by a diffusive mode with flat dispersion, as it typically happens in coherently driven pattern forming systems, such as Benard cells in heat convection \cite{7} or optical parametric oscillators \cite{8}.

The present Letter is devoted to the development of a simple and generic model of a non-equilibrium condensate which does not involve the microscopic physics of the polariton, and can be used to describe the dynamics independently of the details of the specific pumping scheme. Our model is inspired by classical treatments of laser operation \cite{9}, and closely resembles the generic model of atom lasers developed in \cite{10}. In this way, we are able not only to confirm the conclusions of Ref. \cite{6} but also to analytically relate the elementary excitation spectrum to experimentally accessible quantities. The same model is then applied to the Josephson effect \cite{11, 12, 13} in a system of two weakly coupled polaritonic condensates: predictions are given for the frequency and the intrinsic damping rate of Josephson oscillations, and overdamped behavior is anticipated in the case of strong damping.

The experimental scheme used to create the polariton condensate is sketched in Fig.1: under a continuous-wave high energy illumination, hot free carriers are generated in the semiconductor material forming the microcavity. Their cooling down by phonon emission leads the formation of an incoherent gas of bound excitons in the quantum wells, which eventually accumulate in the so-called bottleneck region above the inflection point of the lower exciton-polariton (LP) branch \cite{14}. Polariton-polariton collisions are then responsible for the (generally slower) scattering of polaritons from the bottleneck region to the bottom of the LP branch. For high enough polariton density, Bose stimulation of scattering into the lower part of the LP can take place \cite{15}. When the stimulated scattering rate overcomes losses, the polariton field becomes coherent, and a Bose-Einstein condensate appears \cite{16}.

Our model is based on a mean-field description of the coherent polariton field in terms of a generalized Gross-Pitaevskii equation (GPE) for the condensate macroscopic wavefunction including loss and amplification terms

\begin{equation}
\frac{i}{\hbar} \frac{\partial \psi}{\partial t} = \left\{ \frac{-\hbar \nabla ^2}{2 m_{LP}} + \frac{i}{2} [R(n_R) - \gamma] + g |\psi|^2 + 2 \tilde{g} n_R \right\} \psi.
\end{equation}

As we are interested in the lowest part of the LP dispersion, a parabolic dispersion can be taken for the polaritons, with an effective mass $m_{LP}$. The strength of polariton-polariton interactions within the condensate is fixed by the coupling constant $g$, while $\gamma$ is the polariton damping rate at the bottom of the band. Assuming that relaxation processes are fast enough to ensure local equilibrium in the polariton reservoir in the bottleneck region, and that all coherences between the reservoir and the condensate decay on a fast time-scale compared to the condensate dynamics, the state of the reservoir is fully determined by its polariton density $n_R(x)$. The
amplification rate of the condensate due to stimulated scattering of polaritons from the reservoir is a monotonically growing function \( R(n_R) \) of the reservoir density \( n_R \). Interactions between the condensate and the reservoir polaritons are modelled by the interaction constant \( \tilde{g} \), generally different from the condensate one \( g \). With respect to single-mode theories \[15, 16, 20\], the generalized GPE has the important advantage of fully taking into account the multi-mode nature of the spatially extended polariton field. Moreover, unlike kinetic approaches based on the Boltzmann equation \[15, 16, 20\], the present model is able to describe the coherent dynamics of the condensate. These issues are essential in view of a study of the elementary excitations of the condensate.

The evolution equation \[11\] for the condensate has to be coupled to an equation for the density \( n_R(x) \) of reservoir polaritons:

\[
\frac{\partial n_R}{\partial t} = P - \gamma_R n_R - R(n_R) |\psi(x)|^2 + D \nabla^2 n_R. \tag{2}
\]

Polaritons are pumped in the reservoir with a rate \( P \) and relax at a rate \( \gamma_R \). The spatial hole-burning effect due to the scattering of reservoir polaritons into the condensate is taken into account by the \( R(n_R) |\psi|^2 \) term; spatial diffusion of reservoir polaritons takes place with a diffusion constant \( D \).

The steady state of the system under a continuous-wave and uniform pumping \( P \) can be obtained by substituting the ansatz

\[
\psi(x,t) = e^{-i\mu_{\text{P}} t} \psi_0 \tag{3}
\]

\[
n_R(x,t) = n^0_R. \tag{4}
\]

into \[11\] and \[2\]. For small values of \( P \), no condensate is present \( \psi_0 = 0 \) and the reservoir density is a linear function of the pump intensity \( n^0_R = P/\gamma_R \). This solution is dynamically stable as long as the amplification rate is not able to overcome the losses, i.e. \( R(n^0_R) < \gamma \).

The threshold \( P = P_{th} \) corresponds to the value \( n^0_{th} \) for the reservoir density, which guarantees equilibrium between amplification and losses \( R(n^0_{th}) = \gamma \). When the pumping rate \( P \) is increased above the threshold, the solution \( \psi_0 = 0 \) becomes dynamically unstable and a condensate appears. Stationarity imposes the net gain to vanish, which fixes the reservoir density to the equilibrium value \( n^0_R = n^0_{th} \). The condensate density grows as \( n_c^0 = |\psi_0|^2 = (P - P_{th})/\gamma \), and the oscillation frequency of the macroscopic wavefunction is \( \mu_T = \mu + 2\tilde{g}n^0_c \) with \( \mu = \mu n^0_c \).

As usual \[8, 21, 22\], the elementary excitations spectrum around the stationary state of the system can be obtained by a linearization of the motion equations \[12\] around the steady state solution \[34\]. Thanks to the translational invariance of the system, the fluctuations can be decomposed in their Fourier components:

\[
\psi(x,t) = e^{-i\mu_{\text{P}} t} \psi_0 \left[ 1 + \sum_k u_k e^{i(kr - \omega t)} \right] + \psi^*_k e^{-i(kr - \omega t)} \tag{5}
\]

\[
n_R(t) = n^0_R \left( 1 + w_k e^{i(kr - \omega t)} + w^*_k e^{-i(kr - \omega t)} \right). \tag{6}
\]

Introducing the fluctuation vector \( \mathcal{U}_k = (u_k, v_k, w_k)^T \) and substituting this expansion into the motion equations, one is immediately led to the eigenvalue equation \( \mathcal{L}_k \mathcal{U}_k = \omega \mathcal{U}_k \) defining the elementary excitations, where the generalized Bogoliubov matrix \( \mathcal{L}_k \) is

\[
\mathcal{L}_k = \begin{pmatrix}
\mu + \frac{\hbar k^2}{2m_{\text{L.P}}} & \mu - \frac{\hbar k^2}{2m_{\text{L.P}}} & \frac{i\beta \gamma}{2} + \frac{2\gamma}{\alpha} - \frac{\gamma_R}{\alpha \
\frac{\beta \gamma}{2} & -\mu + \frac{\hbar k^2}{2m_{\text{L.P}}} & \frac{i\beta \gamma}{2} - \frac{2\gamma}{\alpha} - \frac{\gamma_R}{\alpha} \
\frac{\beta \gamma}{2} & -\mu & \frac{i\beta \gamma}{2} + \frac{2\gamma}{\alpha} - \frac{\gamma_R}{\alpha} + i\eta (\gamma_R + D k^2)
\end{pmatrix}
\tag{7}
\]

Here \( \alpha = P/P_{th} - 1 \) is the relative deviation from the threshold pumping intensity and the dimensionless coefficient \( \beta = n^0_{th} R - (n^0_{th}) / R(n^0_{th}) \) characterizes the dependence of the amplification rate on the reservoir density, and \( \eta = 1 + \alpha \beta \). The standard Hartree-Fock value \( \beta = 2g \) has been taken. Quite remarkably, the excitation spectrum does not depend on the actual value of the scattering rate \( R \) of the reservoir into the condensate mode: only the effective exponent \( \beta \) and the relative pumping rate \( \alpha \) do matter. Of course, the threshold value \( P_{th} \) of the pumping intensity does depend (in an inversely proportional way) on \( R \).

![FIG. 1: Top (a) panel: sketch of the pumping and condensate formation scheme. (b-c) panels: real and imaginary part of the excitation spectrum of a homogeneous polariton condensate as a function of the wavevector \( k \) (in units of the healing length \( \xi = \sqrt{\hbar/(m_{\text{L.P}} \mu)} \)). Parameters: \( R/\gamma = 5 \) and \( \alpha = \beta = \gamma/\mu = 1 \) (b,c); \( R/\gamma = 1 \) and \( \alpha = 0.5, \beta = \gamma/\mu = \gamma_R/\mu = 1 \) (d,e). Rescaled diffusion constant \( D m_{\text{L.P}}/\hbar \approx 5 \times 10^{-4} \); the spectra are indistinguishable from the \( D = 0 \) case. Dashed lines in (b,d): Bogoliubov dispersion \[9\] at equilibrium.

Typical examples of the dispersion of elementary excitations around the Bose condensed state are shown in...
whose imaginary part is close to \((1 + \alpha \beta D)\). Physically, this mode can be understood as a slow rotation of the condensate phase across the sample; the generator \((1, -1, 0)^T\) of global phase rotations is indeed an eigenvector of \(\mathcal{L}_{k=0}\) with a vanishing eigenvalue.

Let us analyze the different cases in more detail, starting from the physically most relevant one \(\gamma_R \gg \gamma\) where the state of the reservoir always remains very close to its stationary-state for the given density of condensate polaritons. The dispersion for this case is shown in Fig. 1(b-c): in stark contrast with the linear dispersion of the propagating sound mode in equilibrium Bose-Einstein condensates \([21, 24]\), the Goldstone mode (indicated as + in the figure) shows here a diffusive and non-propagating behavior at low \(k\). The real part is dispersionless and equal to zero, while the imaginary part starts from zero in a quadratic way. This result is in agreement with the prediction of the recent work \([6]\) where these issues were addressed starting from a very specific microscopic model of the polaritons, and suggests that the diffusive behaviour of the Goldstone mode is indeed a generic fact in non-equilibrium phase transitions not only under a coherent pumping as in pattern forming systems \([7, 8]\), but also in the case of incoherent pumping. Note that this diffusive behaviour is in no way due to the spatial diffusion of reservoir polaritons, and would be present even in the absence of spatial diffusion. The value \(D = 5 \text{ cm}^2/\text{s}\) actually chosen in the figures is inspired by recent experimental studies on CdTe quantum well samples \([22]\), and corresponds to a very small value of the dimensionless diffusion constant \(\bar{D} = D m_{LP}/\hbar \approx 5 \times 10^{-4}\).

An analytical explanation of this behavior is readily obtained by adiabatically eliminating the dispersionless and strongly damped reservoir mode \((R\) in the figure) whose imaginary part is close to \((1 + \alpha \beta)\) \(\gamma_R\). Taking for simplicity \(D = 0\), this leads to the following dispersion of the two branches of condensate excitations:

\[
\omega_{\pm}(k) = -\frac{i\Gamma}{2} \pm \sqrt{\omega_{\text{Bog}}(k)^2 - \frac{\Gamma^2}{4}},
\]

where \(\omega_{\text{Bog}}\) is the usual Bogoliubov dispersion of dilute Bose gases at equilibrium

\[
\omega_{\text{Bog}}(k) = \sqrt{\frac{\hbar k^2}{2 m_{LP}^2} + \frac{\hbar^2 k^2}{2 m_{LP}^2} + 2\mu}.
\]

The non-equilibrium nature of the system is quantified by the effective relaxation rate

\[
\Gamma = \alpha \beta \gamma/(1 + \alpha \beta),
\]

whose value tends to 0 when the threshold is approached \(\alpha \gtrsim 0\) and saturates to \(\gamma\) for large \(\alpha \gg 1\). The first + branch of \(\mathfrak{S}\) is the Goldstone branch which corresponds for small \(k\) values to a slow rotation of the condensate phase, while the – branch corresponds to slow modulations of the condensate density; for low \(k\) values, these are damped at the finite rate \(\Gamma\) defined in (10). From \(\mathfrak{S}\) it is immediate to obtain a prediction for the width \(\Delta k\) of the region in \(k\) space in which the dispersion of the Goldstone mode is flat \(\text{Re}[\omega_G(k)] = 0\). Outside this region, i.e. for \(k \gg \Delta k\), the \(\pm\) modes recover the standard Bogoliubov modes of an equilibrium condensate. In order for a sound-like branch to be observable, one however needs that \(\Delta k \ll 1/\xi, \xi = \sqrt{\hbar/(m_{LP} \mu)}\) being the healing length of the condensate. Elementary manipulations show that this condition is actually met if \(\gamma^2 < g \alpha^0_n(1 + \alpha \beta) \gamma_R/\beta\).

More complex features are observed in the case where \(\gamma_R\) and \(\gamma\) have comparable magnitudes \([26]\) and the reservoir can no longer be adiabatically eliminated \([\text{Fig. 1} (\text{d-e})]\).

The most significant feature is the dynamical instability \(\text{Im}[\omega] > 0\) of the DI branch that is visible in panel (e) for \(k \xi \lesssim 1.2\): the homogeneous state \([\mathfrak{S}1]\) is no longer dynamically stable and a spatial modulation has to appear in the steady-state density profile of the condensate. The origin of the instability can be traced back in the repulsive interaction between the condensate and the reservoir polaritons: because of the mean-field interaction potential \(\tilde{g}_{mn}\), a local depletion of the reservoir density \(n_R(z)\) creates a potential well which attracts the condensate \(\psi_2\) values to a slow rotation of the condensate.

As a final point of the Letter, we briefly address the effect of the non-equilibrium condition on the Josephson oscillations between a pair of spatially separated polariton condensates trapped in adjacent wells. Following classical work on the Josephson effect \([11, 12, 21]\), the dynamics can be described by the following set of equations

\[
\frac{d\psi_j}{dt} = -J \psi_{j-1} + U |\psi_j|^2 \psi_j + i \frac{R(n_j - \gamma)}{2} |\psi_j|, \quad j = 1, 2
\]

\[
\frac{d n_j}{dt} = P_j - \gamma_R n_j - R(n_j) |\psi_j|^2
\]

for the amplitude \(\psi_j\) of the two condensates \((j = \{1, 2\})\), and the reservoir densities \(n_j\). These equations can be derived by projecting the GPE onto the localized wavefunctions \(\phi_j\) describing the ground state of each well, and assuming these not to be distorted by the interactions. The total polariton wavefunction then reads: \(\psi(r) = \psi_1 \phi_1(r) + \psi_2 \phi_2(r)\), the charging energy \(U = g \int \sqrt{\phi_j^4}\), and the hopping energy is related to the polariton flux...
atoms per well can be obtained by simply replacing the transitions around the stationary state with a separation between the wells of 1 μm, and a well depth of 3 meV, one obtains that $J = 0.1$ meV and $U = 0.03$ meV for a polariton nonlinearity $g = 0.015$ meV μm$^2$.

The standard normalization $\int d|\phi|^2 = 1$ has been assumed here. Inserting typical values of $2 \mu$m for the well size, a separation between the wells of 1 μm, and a well depth of 3 meV, one obtains that $J = 0.1$ meV and $U = 0.03$ meV for a polariton nonlinearity $g = 0.015$ meV μm$^2$.

Restricting again to the most significant $\gamma_R \gg \gamma$ case, the frequency of the small amplitude Josephson oscillations around the stationary state with $N_{1,2} = |\psi_{1,2}|^2 = N$ atoms per well can be obtained by simply replacing the expression $\omega_J = \sqrt{4J(NU + J)}$ to the Bogoliubov frequency $\omega_{Bog}(k)$ in [5]. Examples of the different regimes for the dynamics around the stationary state are shown in Fig. 2 starting from the steady-state under $P_{1,2} = P_0$, the pumping intensity in each well is modulated to $P_{1,2} = P_0 \pm \Delta P$ for a short time interval $0 < t < T_{exc}$ and then brought back to $P_{1,2} = P_0$. The system dynamics is followed on the mode populations $N_{1,2}$. If the effective damping rate $\Gamma$ is smaller than $\omega_J$ (upper panel), the behaviour closely resembles the usual Josephson oscillations as observed in atomic Bose-Einstein condensates in [13]: the only differences are the intrinsic damping rate $\Gamma$, and the slight frequency shift predicted by [5]. On the other hand, if $\Gamma > \omega_J$ (lower panel), Josephson oscillations are replaced by an exponential relaxation back to the stationary state; the two modes at $\omega_J$ predicted in [5] appear in the relaxation dynamics as two well-separated exponentials.

In summary, we have developed in this Letter a generic model for the coherent dynamics of a polariton Bose-Einstein condensate. The intrinsic non-equilibrium nature of the system is taken into account by means of a generalized Gross-Pitaevskii equation including loss and amplification terms. This model is used to study the elementary excitations around the stationary state of the system. In a spatially homogeneous geometry, a diffusive Goldstone mode is found, and novel features are predicted for the Josephson effect in two-well systems. This formalism will be of great utility in the study of the complex structures which appear in spatially inhomogeneous condensates thanks to the interplay of condensation and losses (see, e.g., 29).

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