Excitations in a Nonequilibrium Bose-Einstein Condensate of Exciton Polaritons

Michiel Wouters[1] and Iacopo Carusotto[2]

[1]*TFVS, Universiteit Antwerpen, Groenenborgerlaan 171, 2020 Antwerpen, Belgium*

[2]*BEC-CNR-INFM and Dipartimento di Fisica, Università di Trento, I-38050 Povo, Italy*

(Received 19 February 2007; published 3 October 2007)

We develop a mean-field theory of the dynamics of a nonequilibrium Bose-Einstein condensate of exciton polaritons in a semiconductor microcavity. The spectrum of elementary excitations around the stationary state is analytically studied by means of a generalized Gross-Pitaevskii equation. A diffusive behavior 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.

DOI: 10.1103/PhysRevLett.99.140402 PACS numbers: 05.30.Jp, 03.75.Kk, 42.65.Sf, 71.36.+c

After a few decades of impressive efforts on a variety of different systems such as bulk cuprous oxide [1] and coupled quantum wells [2], first observations of Bose-Einstein condensation (BEC) of excitons in solid state systems have been recently reported in a gas of exciton polaritons [3] and immediately confirmed by other groups [4–6]. With respect to previous examples of BEC as liquid ^{4}He and ultracold atomic gases, the present system has the crucial novelty of being an intrinsically *nonequilibrium* system: because of the finite lifetime of polaritons, the condensate has 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 balance of pumping and losses. From this point of view, the polariton condensate shares some similarities with a spatially extended laser [7], but a direct analogy is made impossible by the strong nonlinearity due to polariton-polariton collisions. The (still to come) atom laser [8] would be the closest analog of the present polariton BEC.

In addition to its effect on the cloud of noncondensed polaritons [6,9,10], the nonequilibrium condition is responsible for qualitative novelties also in the dynamic behavior of the polariton BEC. The first calculation of the excitation spectrum on top of a homogeneous polariton condensate taking into account its driven-dissipative nature was reported in [11]. As is typical in dynamical systems far from equilibrium such as Bénard cells in heat convection [12] and optical parametric oscillators [13], the lowest-lying excitation mode consists of a diffusive mode instead of a propagating mode like sound. It is the purpose of the present Letter to develop a simple and generic theory of a nonequilibrium condensate which, differently from [11], does not require a microscopic model of the polariton physics, and can be used independently of the specific pumping scheme and in arbitrary geometries. Our theory is inspired by classical treatments of laser operation [14], and closely resembles the generic model of atom lasers developed in [15]. Differently from kinetic approaches based on the Boltzmann equation [9,16–18], our model fully includes the coherence of the polariton field; differently from single-mode theories [19,20], it is able to follow the spatial, i.e., multimode, dynamics of the condensate. Both these features [21] are indeed essential to get a complete description of the coherent dynamics of the condensate. Under reasonable assumptions, our predictions for the diffusive nature of the elementary excitations in a homogeneous system are in perfect agreement with the conclusions of Ref. [11]. As a first application to nontrivial geometries, novel features are anticipated for the Josephson oscillations [22–24] between two weakly coupled polaritonic condensates.

The physical system we consider consists of a semiconductor microcavity containing a few quantum wells with an excitonic transition strongly coupled to the cavity photon mode [3,4]. Its basic excitation modes are exciton polaritons, i.e., linear superpositions of a quantum well exciton and a cavity photon, and satisfy Bose statistics. The experimental scheme used to create the polariton condensate is sketched in Fig. 1(a): 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 to the formation of a 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 [25]. Polariton-polariton collisions are then responsible for the (generally slower) scattering of polaritons from the reservoir in the bottleneck region to the bottom of the LP branch. For high enough polariton density, scattering into the lower part of the LP is enhanced by Bose stimulation effects [9]: when the stimulated scattering rate overcomes losses, the polariton field becomes coherent, and a BEC appears.

Our model is based on a mean-field description of the condensate in terms of a generalized Gross-Pitaevskii equation (GPE) for the macroscopic wave function $\psi(\mathbf{r})$ including loss and amplification terms

$$i\frac{\partial\psi}{\partial t}=\left\{-\frac{\hbar\nabla^2}{2m_{\rm LP}}+\frac{i}{2}[R(n_R)-\gamma]+g|\psi|^2+2\tilde{g}n_R\right\}\psi. \quad (1)$$

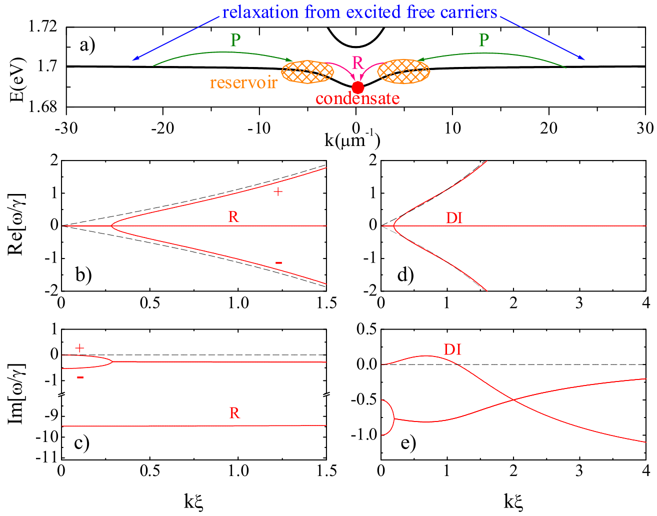

FIG. 1 (color online). Top (a) panel: sketch of the pumping and condensate formation scheme. (b)–(e) panels: real and imaginary part of the excitation spectrum of a homogeneous polariton condensate as a function of the wave vector k (in units of the healing length $\xi = \sqrt{\hbar/m_{\rm LP}\mu}$). Parameters: $\gamma_R/\gamma = 5$ and $\alpha = \beta = \gamma/\mu = 1$ (b),(c); $\gamma_R/\gamma = 1$ and $\alpha = 0.5$, $\beta = \gamma/\mu = \gamma_R/\mu = 1$ (d),(e). Rescaled diffusion constant $Dm_{\rm LP}/\hbar \approx 5 \times 10^{-4}$; the spectra are indistinguishable from the $D = 0$ case. Dashed lines in (b),(d): standard equilibrium Bogoliubov dispersion.

As we are interested in the lowest part of the LP dispersion, the polariton dispersion is approximated by a parabolic one with effective mass $m_{\rm LP}$ and momentum-independent loss rate γ. The strength of polariton-polariton interactions within the condensate is fixed by the coupling constant g. Provided the polariton distribution in the reservoir region and all coherences between the reservoir and the condensate relax on a short time scale as compared to the condensate dynamics, the (non-necessarily thermal) state of the reservoir is fully determined by its local density $n_R(\mathbf{r})$. The amplification rate $R(n_R)$ of the condensate due to stimulated scattering of polaritons from the reservoir is a monotonically growing function of n_R. Interactions between condensate and reservoir polaritons are modeled by the interaction constant $\tilde{g}$, generally different from the condensate one g.

The condensate evolution (1) has to be coupled to an equation for the density $n_R(\mathbf{r})$ of reservoir polaritons. In a simple phenomenological model, this can be written as

$$\frac{\partial n_R}{\partial t} = P - \gamma_R n_R - R(n_R)|\psi(\mathbf{r})|^2 + D\nabla^2 n_R, \quad (2)$$

where polaritons are pumped into the reservoir at a rate P and relax at a rate γ_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; D is the spatial diffusion rate of reservoir polaritons.

The steady state under a continuous-wave and uniform pumping can be obtained by substituting an ansatz of the form $\psi(x,t) = e^{-i\mu_T t}\psi_0$ and $n_R(x,t) = n_R^0$ into (1) and (2). For small values of P, no condensate is present $\psi_0 = 0$ and the reservoir density is proportional to the pump intensity $n_R^0 = P/\gamma_R$. This solution is dynamically stable as long as the amplification is not able to overcome the losses, i.e., $R(n_R^0) < \gamma$. The threshold $P = P_{\rm th}$ corresponds to the value $n_R^{\rm th}$ of the reservoir density which guarantees exact balance of amplification and losses $R(n_R^{\rm th}) = \gamma$. When 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 clamps the reservoir density to $n_R^0 = n_R^{\rm th}$, while the condensate density grows as $|\psi_0|^2 = (P - P_{\rm th})/\gamma$. The oscillation frequency of the macroscopic wave function is $\mu_T = \mu + 2\tilde{g}n_R^0$ with $\mu = g|\psi_0|^2$.

The elementary excitations spectrum around the stationary state of the system can be obtained in the usual way [13,26,27] by linearizing the motion Eqs. (1) and (2) around the steady-state solution. Thanks to the translational invariance, the fluctuations can be decomposed in their Fourier components

$$\psi(\mathbf{r},t) = e^{-i\mu_T t}\psi_0\left[1 + \sum_{\mathbf{k}} u_{\mathbf{k}} e^{i(\mathbf{kr}-\omega t)} + + v_{\mathbf{k}}^* e^{-i(\mathbf{kr}-\omega t)}\right], \quad (3)$$

$$n_R(t) = n_R^0(1 + w_{\mathbf{k}} e^{i(\mathbf{kr}-\omega t)} + w_{\mathbf{k}}^* e^{-i(\mathbf{kr}-\omega t)}). \quad (4)$$

Introducing the fluctuation vector $\mathcal{U}_{\mathbf{k}} = (u_{\mathbf{k}}, v_{\mathbf{k}}, w_{\mathbf{k}})^T$, the eigenvalue equation defining the elementary excitations has the simple matricial form $\mathcal{L}_{\mathbf{k}}\mathcal{U}_{\mathbf{k}} = \omega\mathcal{U}_{\mathbf{k}}$ with

$$\mathcal{L}_{\mathbf{k}} = \begin{pmatrix} \mu + \frac{\hbar k^2}{2m_{\rm LP}} & \mu & \frac{i\beta\gamma}{2} + \frac{2\gamma\mu}{\alpha\gamma_R} \\ -\mu & -\mu - \frac{\hbar k^2}{2m_{\rm LP}} & \frac{i\beta\gamma}{2} - \frac{2\gamma\mu}{\alpha\gamma_R} \\ -i\alpha\gamma_R & -i\alpha\gamma_R & -i[\eta\gamma_R + Dk^2] \end{pmatrix}. \quad (5)$$

Here $\alpha = P/P_{\rm th} - 1$ is the relative deviation from the threshold pumping intensity and the dimensionless coefficient $\beta = n_R^0 R'(n_R^0)/R(n_R^0)$ characterizes the dependence of the amplification rate on the reservoir density, and $\eta = 1 + \alpha\beta$. The standard Hartree-Fock value $\tilde{g} = 2g$ 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 β and the relative pumping rate α do matter. Of course, the threshold value $P_{\rm th}$ of the pumping intensity does depend (in an inversely proportional way) on R.

Typical examples of elementary excitation spectra are shown in Figs. 1(b)–1(d). As Bose condensation corresponds to a spontaneous breaking of a $U(1)$ symmetry, all spectra involve a Goldstone branch whose dispersion $\omega_G(k)$ tends to 0 in the long-wavelength $k \to 0$ limit [12,26]. 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 $\mathcal{L}_{\mathbf{k}=0}$ with a vanishing eigenvalue.

Let us now analyze in more detail the different cases, starting from the simplest, yet physically most relevant one $\gamma_R \gg \gamma$ [28] when the reservoir is able to adiabatically follow the evolution of the condensate. The dispersion of elementary excitations is shown in Figs. 1(b) and 1(c): in stark contrast with the linear dispersion of the propagating sound mode in equilibrium Bose-Einstein condensates [26], the Goldstone mode (indicated as + in the figure) shows a diffusive and nonpropagating 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 conclusion is in agreement with Ref. [11] where these issues were investigated using a very specific microscopic model of the polaritons. This suggests that the diffusive behavior of the Goldstone mode is a generic fact of non-equilibrium phase transitions not only under a coherent pumping as in pattern forming systems [12,13], but also in the present case of incoherent pumping. Note that this diffusive behavior is in no way due to the spatial diffusion of reservoir polaritons, and would be observed even in its absence. The value $D = 5\ \mathrm{cm}^2/\mathrm{s}$ actually chosen in the figures is inspired by recent experimental studies [29] and corresponds to a very small dimensionless diffusion constant $\tilde{D} = D m_{\mathrm{LP}}/\hbar \approx 5 \times 10^{-4}$.

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:

$$\omega_\pm(k) = -i\Gamma/2 \pm \sqrt{[\omega_{\mathrm{Bog}}(k)]^2 - \Gamma^2/4}, \qquad (6)$$

where $\omega_{\mathrm{Bog}}(k)$ is the usual Bogoliubov dispersion of dilute Bose gases at equilibrium $\omega_{\mathrm{Bog}}(k) = [\varepsilon_k(\varepsilon_k + 2\mu)]^{1/2}$ and $\varepsilon_k = \hbar k^2/2m_{\mathrm{LP}}$. The nonequilibrium 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 γ for large $\alpha \gg 1$. The first + branch of (6) is the Goldstone branch which corresponds for small k values to a slow rotation of the condensate phase. The − branch corresponds instead to modulations of the condensate density; for low k values, it is damped at a finite rate Γ. From (6) it is immediate to obtain the width Δk of the k-space region where the Goldstone mode is flat $\mathrm{Re}[\omega_G(k)] = 0$. Inserting parameters ($\hbar\Gamma = 0.8$ meV, $m_{\mathrm{LP}} = 10^{-4} m_e$, $\hbar\mu = 0.5$ meV) inspired by the recent experiment [3], one finds that the Goldstone mode is dispersionless down to wavelengths of the order of 10 μm, a value that fits well within the present condensate sizes. On the other hand, for $k \gg \Delta k$, the ± modes recover the standard Bogoliubov dispersion of an equilibrium condensate.

More complex behaviors are predicted if comparable values are taken for γ_R and γ. In this case, the reservoir mode takes full part in the system dynamics and is strongly mixed with the condensate ones. As it is shown in Fig. 1(d) and 1(e), this can result in a dramatically different dispersion of the elementary excitations, note, in particular, the dynamical instability $\mathrm{Im}[\omega] > 0$ of the DI branch for $k\xi \lesssim 1.2$. The origin of this phenomenon is due to the repulsive interactions between condensate and reservoir polaritons. A local depletion of the reservoir density $n_R(\mathbf{r})$ creates a potential well which attracts the condensate polaritons. This in turn leads to a further drop of the local reservoir density by the hole-burning effect, until a spatially modulated steady-state is eventually reached.

As a final point of the Letter, we now discuss the peculiar nonequilibrium features of the Josephson effect between a pair of spatially separated polariton condensates in a two-well geometry. The experimental realization of such a system appears to be feasible in the next future: as demonstrated in [30], polariton traps of arbitrary shape and size can be created with photolithographic techniques. Following classical work [22,23,26], the theoretical description of the two-well system can be simplified by projecting (1) onto the localized ground-state wave functions $\phi_{1,2}$ of each well (normalized as usual as $\int d\mathbf{r}|\phi_j|^2 = 1$). In terms of the amplitudes $\psi_{1,2}$ in the two wells, the total polariton wave function reads $\psi(\mathbf{r}) = \psi_1\phi_1(\mathbf{r}) + \psi_2\phi_2(\mathbf{r})$, and the dynamics is

$$i\frac{d\psi_j}{dt} = -J\psi_{3-j} + U|\psi_j|^2\psi_j + \frac{i}{2}[R(n_j) - \gamma]\psi_j, \qquad (7)$$

$$\frac{dn_j}{dt} = P_j - \gamma_R n_j - R(n_j)|\psi_j|^2. \qquad (8)$$

Here, $n_{1,2}$ are the reservoir densities at the two condensate positions, the charging energy is $U = g\int d\mathbf{r}|\phi_j|^4$, and the hopping energy J is related to the polariton flux through a surface separating the wells by [26]: $J = (\hbar/m_{\mathrm{LP}}) \times \int_{z=0} d\sigma(\phi_1^*\partial_z\phi_2 - \phi_2^*\partial_z\phi_1)$. As a specific example, we have considered a double-well geometry inspired from the recent experiments [30], i.e., a pair of circular wells of depth of 3 meV and radius 2 μm, separated by 1 μm. The resulting values $J = 0.1$ meV and $U = 0.03$ meV (for

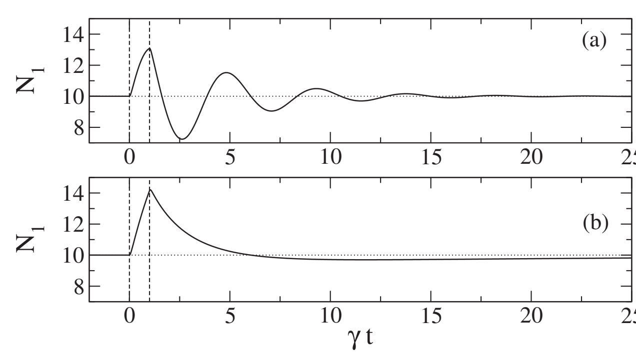

FIG. 2. Time evolution of the population N_1 after an excitation sequence of duration $\gamma T_{\mathrm{exc}} = 1$ (dashed vertical lines). Upper (a) panel: Josephson oscillations ($J = \gamma/2$). Lower (b) panel: overdamped relaxation ($J = \gamma/20$). Other parameters: $U = J/10$, $\gamma_R = 10\gamma$, $R_0 = \gamma$, $P_0/P_{\mathrm{thr}} = 2$, $\Delta P/P_0 = 0.5$.

$g = 0.015$ meV μm^2) are quite promising in view of experimental investigations.

Restricting ourselves to the most significant $\gamma_R \gg \gamma$ case, the frequency of the small amplitude Josephson oscillations around the stationary state with $N_{1,2} = |\psi_{1,2}|^2 = N$ polaritons per well can be obtained by simply replacing the expression $\omega_J = \sqrt{4J(NU + J)}$ to the Bogoliubov frequency $\omega_{\rm Bog}(k)$ in (6). Examples of the different regimes 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 $0 < t < T_{\rm exc}$ and then brought back to $P_{1,2} = P_0$. The system dynamics is followed on the mode populations $N_{1,2}$. If $\omega_J > \Gamma$ (upper panel), the only difference as compared to Josephson oscillations in atomic BECs [24] consists of the intrinsic damping rate Γ. 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 $\omega_\pm$ appear in the relaxation dynamics as two well-separated exponentials.

To summarize, we have developed a generic model for the dynamics of a polariton Bose-Einstein condensate. The intrinsic nonequilibrium nature of the system is taken into account by means of a generalized Gross-Pitaevskii equation including loss and amplification terms. The dispersion of elementary excitations around the stationary state is investigated in the different regimes: under a reasonable fast reservoir hypothesis, the Goldstone mode of a spatially homogeneous system is found to be diffusive and the Josephson oscillations in a two-well system overdamped. This model will be of great utility to study the complex structures appearing [31] in spatially inhomogeneous condensates under the combined effect of condensation and losses, and will be the starting point for studies of the critical properties of the BEC phase transition in a nonequilibrium context.

Continuous stimulating discussions with A. Baas, C. Ciuti, M. Richard, D. Sarchi, M. H. Szymańska, and V. Savona are warmly acknowledged. This research has been supported financially by the FWO-V Projects No. G.0356.06, No. G.0115.06 and the Special Research Fund of the University of Antwerp, BOF.NOI UA 2004. M. W. acknowledges financial support from the FWO-Vlaanderen in the form of a "mandaat postdoctoraal onderzoeker." I. C. acknowledges hospitality at the Centre Emile Borel of the Institut Henri Poincaré and financial support from CNRS.

[1] J. I. Jang and J. P. Wolfe, Phys. Rev. B **72**, 241201(R) (2005); D. Fröhlich *et al.*, Phys. Status Solidi B **243**, 2367 (2006); K. Yoshioka and M. Kuwata-Gonokami, Phys. Rev. B **73**, 081202 (2006).
[2] L. V. Butov, J. Phys. Condens. Matter **19**, 295202 (2007); Z. Vörös, D. W. Snoke, L. Pfeiffer, and K. West, Phys. Rev. Lett. **97**, 016803 (2006).
[3] J. Kasprzak *et al.*, Nature (London) **443**, 409 (2006).
[4] H. Deng *et al.*, Phys. Rev. Lett. **97**, 146402 (2006).
[5] S. Christopoulos *et al.*, Phys. Rev. Lett. **98**, 126405 (2007).
[6] R. Balili *et al.*, Science **316**, 1007 (2007).
[7] K. Staliunas and V. Sanchez-Morcillo, *Transverse Patterns in Nonlinear Optical Resonators* (Springer, New York, 2003).
[8] K. Helmerson *et al.*, Phys. World **12**, 31 (1999); A. P. Chikkatur *et al.*, Science **296**, 2193 (2002); T. Lahaye *et al.*, Phys. Rev. Lett. **93**, 093003 (2004).
[9] D. Porras *et al.*, Phys. Rev. B **66**, 085304 (2002).
[10] D. Sarchi and V. Savona, Phys. Rev. B **75**, 115326 (2007).
[11] M. H. Szymańska, J. Keeling, and P. B. Littlewood, Phys. Rev. Lett. **96**, 230602 (2006).
[12] M. C. Cross and P. C. Hohenberg, Rev. Mod. Phys. **65**, 851 (1993).
[13] M. Wouters and I. Carusotto, arXiv:cond-mat/0606755 [Phys. Rev. A (to be published)].
[14] W. Lamb, Phys. Rev. **134**, A1429 (1964).
[15] B. Kneer *et al.*, Phys. Rev. A **58**, 4841 (1998).
[16] F. Tassone and Y. Yamamoto, Phys. Rev. B **59**, 10830 (1999).
[17] T. D. Doan *et al.*, Phys. Rev. B **72**, 085301 (2005).
[18] G. Malpuech *et al.*, Phys. Rev. B **65** , 153310 (2002).
[19] P. Schwendimann and A. Quattropani, Phys. Rev. B **74**, 045324 (2006).
[20] Y. G. Rubo *et al.*, Phys. Rev. Lett. **91**, 156403 (2003); F. P. Laussy *et al.*, Phys. Rev. Lett. **93**, 016402 (2004).
[21] An attempt to solve these same issues was made in [10] by combining the kinetic theory with a quasistationary Bogoliubov description of the condensate. Yet, no predictions were made for the very nonequilibrium features of the excitation spectrum of the system.
[22] G. J. Milburn *et al.*, Phys. Rev. A **55**, 4318 (1997).
[23] A. Smerzi *et al.*, Phys. Rev. Lett.**79**, 4950 (1997); S. Giovanazzi *et al.*, Phys. Rev. Lett. **84**, 4521 (2000).
[24] M. Albiez *et al.*, Phys. Rev. Lett. **95**, 010402 (2005).
[25] F. Tassone *et al.*, Phys. Rev. B **56**, 7554 (1997).
[26] L. P. Pitaevskii and S. Stringari, *Bose-Einstein Condensation* (Clarendon Press, Oxford, 2003).
[27] I. Carusotto and C. Ciuti, Phys. Rev. Lett. **93**, 166401 (2004); C. Ciuti and I. Carusotto, Phys. Status Solidi B **242**, 2224 (2005).
[28] Although the lifetime of bottleneck polaritons is comparable to the one of condensate polaritons, available work suggest that a much larger γ_R should be used to effectively model the rapid relaxation within the reservoir.
[29] M. T. Portella-Oberli *et al.*, Phys. Rev. B **66**, 155305 (2002).
[30] O. El Daïf *et al.*, Appl. Phys. Lett. **88**, 061105 (2006); R. Idrissi Kaitouni *et al.*, Phys. Rev. B **74**, 155311 (2006).
[31] M. Richard *et al.*, Phys. Rev. Lett. **94**, 187401 (2005).