Observation of Long-Lived Polariton States in Semiconductor Microcavities across the Parametric Threshold

D. Ballarini,[1] D. Sanvitto,[1] A. Amo,[1] L. Viña,[1] M. Wouters,[2] I. Carusotto,[3] A. Lemaitre,[4] and J. Bloch[4]

[1]*Departamento Física de Materiales, Universidad Autonóma de Madrid, 28049 Madrid, Spain*

[2]*Institute of Theoretical Physics, Ecole Polytechnique Fédérale de Lausanne EPFL, CH-1015 Lausanne, Switzerland*

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

[4]*LPN/CNRS, Route de Nozay, 91460, Marcoussis, France*

(Received 21 July 2008; revised manuscript received 20 December 2008; published 2 February 2009)

The excitation spectrum around the pump-only stationary state of a polariton optical parametric oscillator in semiconductor microcavities is investigated by time-resolved photoluminescence. The response to a weak pulsed perturbation in the vicinity of the idler mode is directly related to the lifetime of the elementary excitations. A dramatic increase of the lifetime is observed for a pump intensity approaching and exceeding the optical parametric oscillator threshold. The observations can be explained in terms of a critical slowing down of the dynamics upon approaching the threshold and the following appearance of a soft Goldstone mode in the spectrum.

DOI: 10.1103/PhysRevLett.102.056402 PACS numbers: 71.36.+c, 78.45.+h, 78.55.Cr

One of the most striking consequences of the quantum nature of matter is Bose-Einstein condensation (BEC), a phase transition that does not depend on interactions between the microscopic constituents but only on their indistinguishability. So far, BEC has been investigated mostly in equilibrium or quasiequilibrium systems such as liquid helium and trapped ultracold atomic gases [1], but recent developments in solid state physics [2,3] are opening the way towards the study of BEC in nonequilibrium contexts where the state of the many particle systems is no longer determined by a thermodynamical equilibrium condition, but rather by a balance between the external driving and dissipation [4].

Here, we consider the case of exciton-polaritons in semiconductor microstructures: the mixed excitonic and photonic nature of polaritons allows in fact for a number of remarkable properties, e.g., a very light mass, significant interactions, and the possibility of all-optical manipulation and diagnostics [5]. Several groups have reported the observation of spontaneous coherence of polaritons with mechanisms that can be interpreted as nonequilibrium analogs of BEC [2]. In spite of the different pumping schemes (resonant for the optical parametric oscillator—OPO—case [6,7], nonresonant for the so-called polariton BEC case [8]), a $U(1)$ symmetry is spontaneously broken in all cases, and coherence is not simply inherited from the pump beam. Away from the bistability regime [9], the OPO transition is smooth and shows a critical behavior that closely resembles the one of a second-order-like phase transition [10]. As a continuous symmetry is spontaneously broken above the transition point, the lifetime of elementary excitations remains divergent in the long wavelength limit in agreement with the Goldstone theorem of statistical mechanics [11,12].

In the present Letter, we experimentally investigate and theoretically model the lifetime of the elementary excitations of a quantum fluid of polaritons as the pump intensity is spanned across the threshold in the OPO configuration. The steady state of the system is probed by injecting extra polaritons by means of a weak pulsed beam, and the decay time of the response is measured as a function of the pump intensity. A dramatic slowing down of the dynamics is observed as the threshold is approached from below: close to the threshold, the decay time can become orders of magnitude longer than the typical lifetime of polaritons, and it remains very long even well above the threshold. Good agreement between the experimental observations and the theoretical model based on the generalized polariton Gross-Pitaevskii equation is found. Our observation suggests the possibility of investigating the polariton dynamics beyond the limits imposed by the intrinsic polariton lifetime [13].

The experiments are performed at 10 K on a microcavity sample [14] with Rabi splitting of $2\Omega_R = 4.4$ meV and at zero detuning between cavity mode and exciton state. In the OPO configuration, polaritons are coherently injected by a pump beam, which resonantly populates a polariton mode with a defined momentum and energy (pump state). Our pump beam is a continuous-wave laser ($Ti{:}Al_2O_3$), which excites the sample with an incident angle of 10° and has a 45 μm spot diameter. Its frequency is chosen close to resonance with the lower polariton branch (LPB) to inject polaritons with a given wave vector k_p and energy $\hbar\omega_p$. Polariton-polariton collisions are responsible for the parametric scattering of pump polaritons into a pair of signal and idler modes. The efficiency of the scattering was optimized by tuning the pump at a frequency $\hbar\omega_P = 1.5273$ eV, slightly above the linear-regime LP dispersion [9,15], $\varepsilon_{\mathrm{LPB}}(k_P)$, $\hbar\omega_p - \varepsilon_{\mathrm{LPB}}(k_P) \sim \gamma(k_p)$. Here $\gamma(k_p) \sim 0.4$ meV is the LPB linewidth at k_p, while the laser linewidth is $\sim$0.1 meV. An additional 2-ps-long probe pulse is incident on the sample with a tunable angle and is focused within the pump spot with a smaller, 25 μm, spot diameter. The power of the probe is kept low enough to be in the linear-response regime. Photoluminescence (PL) is collected and analyzed by a spectrograph coupled either to a streak- or a conventional CCD camera.

First, we have investigated the stationary-state polariton emission in the absence of the probe. Typical energy-momentum emission patterns are shown in Fig. 1 for two different values of the cw pump power I_p. Although k_p lies well outside the k-space region imaged in Fig. 1, a small polariton occupation of the LPB bottom still appears as a consequence of incoherent relaxation processes even in the linear regime (at low pump powers, lower panel). At higher pump intensities, polariton-polariton interactions are able to significantly modify the emission pattern and, in particular, are responsible for parametric processes, where two pump polaritons at k_p are transformed into a pair of signal and idler polaritons of wave vectors $k_{s,i}$, respectively. The onset of parametric oscillation is clearly visible in the emission for pump intensities above the threshold $I_p^{\rm th} = 12.5$ mW [Fig. 1(a)]: the occupation of the signal at $k_s \simeq 0$ becomes very large and the linewidth of the emission in energy is substantially reduced as compared to the bare LPB linewidth shown in Fig. 1(b). The spectral narrowing in energy is accompanied by a significant broadening in k space. The flat shape of the coherent OPO emission in the $(k_x; E)$ plane is, however, more likely to be a consequence of the peculiar shape of finite-size nonequilibrium condensates discussed in [16] rather than evidence of the diffusive nature of the Goldstone mode [17].

To clearly identify the threshold, we have studied the energy of the signal emission as a function of pump power (triangles in Fig. 2). A smooth and almost linear blueshift of the signal energy is visible at low powers, while a sudden jump appears for I_p just above 12 mW due to the onset of parametric oscillation. Such a discontinuous behavior around the threshold was predicted in Refs. [9,15]. Far above threshold, the blueshift saturates.

The main object of our study is the response of the system in its stationary state to an additional weak ($\lesssim$0.2 mW) probe pulse that impinges on the sample at a large angle of around 20°, i.e., in the vicinity of the idler. The system response to the probe pulse is monitored by investigating the time- and momentum-resolved signal emission and, in particular, its decay time. As an example, we have traced in the inset of Fig. 2 the time evolution of the difference $\Delta I_s = I_s(\text{pump} + \text{probe}) - I_s(\text{pump})$ between the signal emission intensity with and without probe, respectively, for three different pump intensities.

Right after the arrival of the probe pulse, parametric scattering of pump polaritons into k_s is stimulated by the small population of the new idler polaritons injected by the probe (see the fast switch-on of ΔI_s in the inset in Fig. 2). The fast decay on a 30 ps scale is followed by a much slower exponential decay on a time scale in the 100 ps range, i.e., orders of magnitude longer than the empty cavity decay time (2 ps) and the polariton-polariton scattering time [18], but still significantly shorter than the probe repetition time of ~12 ns.

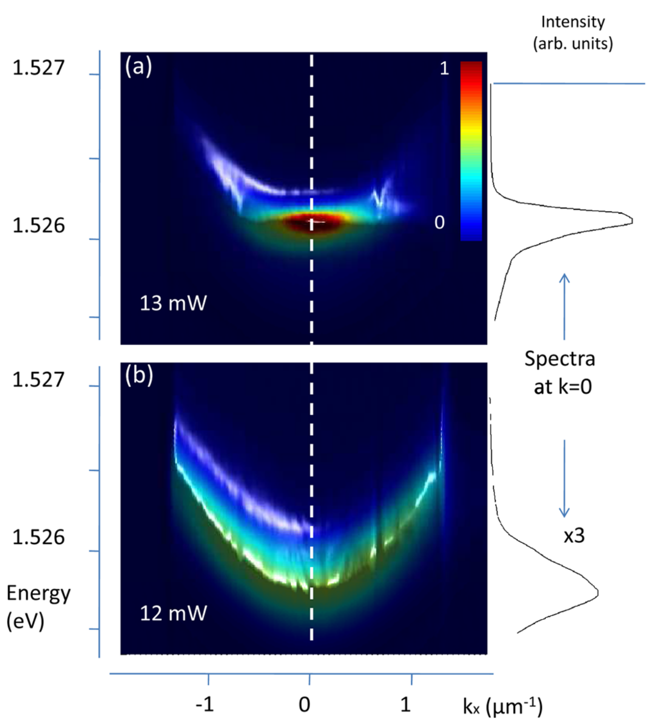

FIG. 1 (color online). Direct experimental observation with the CCD camera of a two-dimensional section $(k_x; E)$, with $k_y = 0$, of the lower polariton dispersion for pump powers I_p (a) just above (13) and (b) below (12 mW) the OPO threshold $I_p^{\rm th} = 12.5$ mW. Energy spectra at $k_x = 0$ (dashed line in the figure) are depicted on the right, with a magnification of factor 3 for the lower panel of the figure. The PL emission is normalized to 1 and plotted in a linear color scale.

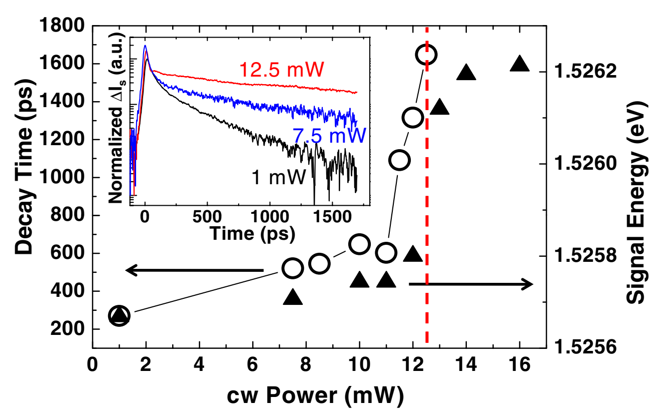

FIG. 2 (color online). Energy (triangles) and decay time (open circles) of the signal versus pump power. The red dashed line indicates the pump threshold. Inset: time evolution of the energy-integrated signal emission ΔI_s for three pump powers (1 mW, 7.5 mW, 12.5 mW).

The response of the system strongly depends on I_p. This dependence of the long decay time is summarized by the circles in Fig. 2. The decay time shows a divergent behavior [19] approaching $I_p = 12.5$mW; for higher powers, it exceeds the time window of our setup. The parametric nature of the enhanced lifetime is confirmed by the coincidence of the divergence with the signal energy jump and the PL frequency narrowing (see Fig. 1).

The role of the parametric processes is further evidenced by the momentum-resolved data shown in Fig. 3. The considered wave vector range is centered around the value k_s where signal emission would appear if the pump intensity was above threshold. While the decay time is a smooth function of k_x for I_p well below the parametric oscillation threshold (open circles), a marked peak is apparent in the vicinity of k_s ($k_x \approx 0$) for pump intensities around and above the threshold (filled circles).

A convenient and physically meaningful way to interpret the slowing down of the response to the probe is to use the coherent polariton model based on a pair of coupled Gross-Pitaevskii-like nonlinear wave equations for the photon and exciton fields $\psi_{C,X}(\mathbf{r}, t)$ [20,21],

$$i\frac{\partial \psi_C}{\partial t} = \left[\omega_C(-i\nabla_{\mathbf{r}}) - i\frac{\gamma_C}{2}\right]\psi_C + \Omega_R\psi_X + F(\mathbf{r}, t), \quad (1)$$

$$i\frac{\partial \psi_X}{\partial t} = \left[\omega_X\psi_X - i\frac{\gamma_X}{2}\right]\psi_X + \Omega_R\psi_C + g|\psi_X|^2\psi_X. \quad (2)$$

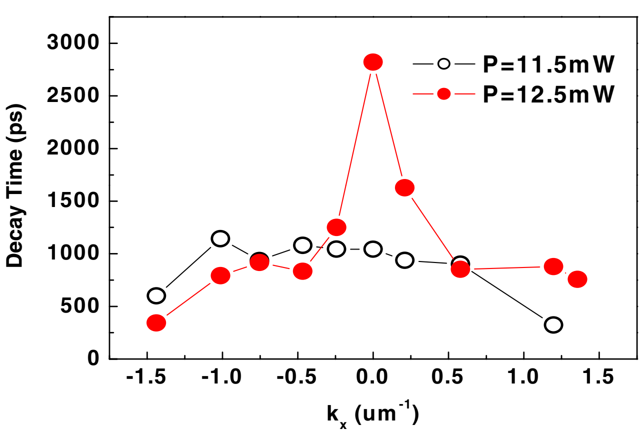

FIG. 3 (color online). k_x dependence of the signal decay time for two different pump powers, 11.5 mW (open circles) and 12.5 mW (filled circles). The high values of the decay time (>500 ps) far from $k_x = 0$ are due to the integration over all the spot area and over all the emitted energies: the intensity profile (not shown) for both 11.5 mW and 12.5 mW is strongly peaked in the parametric scattering region around the signal state at $k_x \simeq 0$, while a much weaker and slower incoherent emission coming from higher energy states (e.g,. residual excitons) and from the border of the spot is responsible for the tails at $|k_x| > 0.5\ \mu$m^{-1}.

We follow the dynamics of the system starting from the $\psi_{X,C}(\mathbf{r}, t) = 0$ vacuum state. $\omega_C(\mathbf{k})$ is the photon dispersion, while the exciton dispersion is assumed to be flat at ω_X. $\gamma_{C,X}$ are the decay rates of the cavity photon and the exciton, respectively. The exciton-exciton interactions are characterized by the nonlinear coupling coefficient g, and Ω_R is the exciton-photon Rabi coupling. The driving $F(\mathbf{r}, t)$ is proportional to the incident electromagnetic field and has to include both the cw pump and the pulse probe: once the system has attained its stationary state under the cw pump only, an additional short probe pulse is applied close to resonance with the idler. The following response of the system is monitored on the most relevant observables, in particular, the polariton distribution in k space. For the sake of simplicity, we have limited ourselves to the case of a plane-wave pump with a well-defined wave vector k_p and periodic boundary conditions, while the finite spatial size of the probe beam is fully taken into account.

As discussed in Refs. [9,21], the approach to the OPO threshold from below is signaled by the decay rate of some mode tending to zero. As a function of k, the decay time results then strongly peaked around the point where the decay rate is the smallest: the closer to threshold, the higher the peak value. These general claims are perfectly visible in the numerical result plotted in Fig. 4. For pump intensities just below $I_p^{\rm th}$, the evolution of the inte-

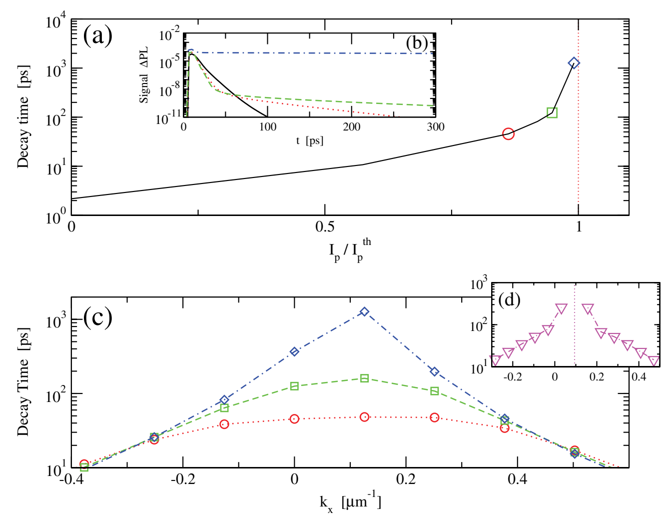

FIG. 4 (color online). Results of numerical calculations. (a) Decay time of ΔI_s as a function of cw pump intensity. Inset (b): Time dependence of k-integrated signal emission ΔI_s for different values of $I_p/I_p^{\rm th} = 5 \times 10^{-4}$ (solid line), 0.57 (dotted line), 0.95 (dashed line), 0.99 (dash-dotted line); integration is performed over the k-space region surrounding k_s. (c) k dependence of the decay time for different values of $I_p/I_p^{\rm th} =$ 0.57, 0.95, 0.99 below threshold. Inset (d): k dependence of the decay time for $I_p/I_p^{\rm th} = 1.15$, above threshold; the vertical dotted line indicates the coherent signal emission wave vector k_s, at which the decay time is infinite by definition of spontaneous symmetry breaking.

grated signal emission $\Delta I_s(t)$ after the arrival of the probe pulse is characterized by a short transient followed by a much slower exponential decay, with a time constant that dramatically increases as the threshold is approached [inset (b)]. By comparing the overall decay time of the integrated $\Delta I_s(t)$ [panel (a)] with the k-dependent decay time [panel (c)], it can immediately be seen that the former is determined by the decay time of the longest-lived mode, a quantity that increases in magnitude and becomes progressively more peaked as the threshold is approached.

This theoretical result is in good agreement with the experimental observations for pump intensities in the vicinity of the threshold, but some specific attention has to be paid to the experimental data for very low pump power. In this regime, the theoretical calculations predict that the decay time should go back to the bare polariton lifetime, while a quite long decay time is observed in the experiment. To explain this behavior, one can mention the spatial inhomogeneity of the system as well as the presence of residual excitons in long-lived states that relax down on a long time scale. These incoherent scattering processes are most important for low powers, while coherent parametric processes take it over as the threshold is approached.

The decay time above threshold is too long to be quantitatively measured with our setup. Numerical simulations do not suffer from such a difficulty, and we summarize here the main features that one expects for this regime. As a consequence of the spontaneous breaking of the $U(1)$ symmetry corresponding to simultaneous rotations of the signal/idler phases, the spectrum of the elementary excitations is characterized by the presence of a soft Goldstone mode corresponding to slow twists in space of the signal/idler phases: as the wave vector $q = k - k_s$ of the excitation tends to zero, both its frequency and decay rate tend to zero [17]. This prediction is confirmed by the numerical results for the k-dependent decay rate shown in Fig. 4(d): the peaked structure of the decay rate as a function of $k - k_s$ is apparent.

In conclusion, we have investigated the response of a continuously pumped OPO to an additional weak pulse in the vicinity of the idler. The emission from the signal is resolved in time and its momentum distribution is analyzed as a function of the pump intensity: when the pump intensity is close or above the parametric oscillation threshold, a lifetime orders of magnitude longer than the intrinsic polariton lifetime is observed for polariton modes in the vicinity of the signal emission. The experiments are explained in terms of a critical slowing down of the elementary excitation dynamics as the threshold is approached. The use of a long living coherent polariton state is of fundamental interest to study, for instance, quantum dynamics of nonequilibrium condensates as shown in Ref. [13].

This work was partially supported by the Spanish Ministerio de Educacion y Ciencia (MAT2005-01388, NAN2004-09109-C04-04, and QOIT-CSD2006-00019), the Comunidad Autonoma de Madrid (S-0505/ESP-0200), and IMDEA-Nanociencia. D. S. thanks the Ramon y Cajal Programme. I. C. is indebted to C. Ciuti for continuous discussions and acknowledges support from the Italian MIUR, the French CNRS and IFRAF, and the EuroQUAM-FerMix program.

[1] D. Pines and P. Nozieres, *The Theory of Quantum Liquids* (Addison-Wesley, Redwood City, 1966), Vols. 1 and 2; L. Pitaevskii and S. Stringari, *Bose—Einstein Condensation* (Oxford University Press, Oxford, 2003).
[2] J. Keeling, F. M. Marchetti, M. H. Szymanska, and P. B. Littlewood, Semicond. Sci. Technol. **22**, R1 (2007).
[3] S. O. Demokritov, *et al.*, Nature (London) **443**, 430 (2006).
[4] *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz, Statistical Mechanics of Driven Diffusive Systems (Academic, NY, 1989), Vol. 17.
[5] Special issue on Microcavities, edited by J. J. Baumberg and L. Viña [Semicond. Sci. Technol. **18**, 10 (2003)].
[6] R. M. Stevenson *et al.*, Phys. Rev. Lett. **85**, 3680 (2000); J. J. Baumberg *et al.*, Phys. Rev. B **62**, R16 247 (2000); R. Houdre, C. Weisbuch, R. P. Stanley, U. Oesterle, and M. Ilegems, Phys. Rev. Lett. **85**, 2793 (2000); A. Baas, J. P. Karr, M. Romanelli, A. Bramati, and E. Giacobino, Phys. Rev. Lett. **96**, 176401 (2006).
[7] N. A. Gippius *et al.*, Europhys. Lett. **67**, 997 (2004); D. N. Krizhanovskii *et al.*, Phys. Rev. B **77**, 115336 (2008).
[8] J. Kasprzak *et al.*, Nature (London) **443**, 409 (2006); H. Deng, G. S. Solomon, R. Hey, K. H. Ploog, and Y. Yamamoto, Phys. Rev. Lett. **99**, 126403 (2007); R. Balili *et al.*, Science **316**, 1007 (2007).
[9] M. Wouters and I. Carusotto, Phys. Rev. B **75**, 075332 (2007).
[10] I. Carusotto and C. Ciuti, Phys. Rev. B **72**, 125335 (2005).
[11] K. Huang, *Statistical Mechanics* (Wiley, New York, 1963).
[12] M. C. Cross and P. C. Hohenberg, Rev. Mod. Phys. **65**, 851 (1993).
[13] A. Amo *et al.*, Nature (London) **457**, 291 (2009).
[14] M. Perrin, P. Senellart, A. Lemaitre, and J. Bloch, Phys. Rev. B **72**, 075340 (2005).
[15] D. M. Whittaker, Phys. Rev. B **71**, 115301 (2005).
[16] M. Wouters, I. Carusotto, and C. Ciuti, Phys. Rev. B **77**, 115340 (2008).
[17] M. Wouters and I. Carusotto, Phys. Rev. A **76**, 043807 (2007).
[18] A. Huynh, *et al.*, Phys. Rev. Lett. **90**, 106401 (2003).
[19] Such long-time features were not accessible to frequency-domain luminescence experiments. See, e.g., P. G. Savvidis, *et al.*, Phys. Rev. B **64**, 075311 (2001).
[20] C. Ciuti, P. Schwendimann, and A. Quattropani, Semicond. Sci. Technol. **18**, S279 (2003).
[21] C. Ciuti and I. Carusotto, Phys. Status Solidi (b) **242**, 2224 (2005).