Polariton Laser Using Single Micropillar GaAs-GaAlAs Semiconductor Cavities

Daniele Bajoni, Pascale Senellart, Esther Wertz, Isabelle Sagnes, Audrey Miard, Aristide Lemaître, and Jacqueline Bloch[*]

CNRS-Laboratoire de Photonique et Nanostructures, Route de Nozay, 91460 Marcoussis, France

Polariton lasing is demonstrated on the zero-dimensional states of single GaAs/GaAlAs micropillar cavities. Under nonresonant excitation, the measured polariton ground-state occupancy is found as large as 10^4. Changing the spatial excitation conditions, competition between several polariton lasing modes is observed, ruling out Bose-Einstein condensation. When the polariton state occupancy increases, the emission blueshift is the signature of self-interaction within the half-light half-matter polariton lasing mode.

Boson statistics can lead to massive occupation of a single quantum state and trigger final state stimulation. This stimulation is responsible for the bright coherent emission of light in a laser. Another fascinating property of massive bosons in thermal equilibrium is their ability to accumulate in the lowest energy state under a given critical temperature. First predicted in 1925 [1], Bose-Einstein condensation was achieved in 1995 for ultracold atoms [2,3]. Demonstrating such bosonic effects with matter waves in a solid-state system is interesting both from a fundamental point of view and for applications since it could provide a new source of coherent light. Cavity polaritons are an example of quasiparticles behaving as bosons at low density [4,5]. They are the exciton-photon mixed quasiparticles arising from the strong coupling regime between quantum well (QW) excitons and a resonant optical cavity mode. Because of their small effective mass (10^{-8} times that of the hydrogen atom) cavity polaritons are expected to condensate at unusually high temperatures (up to room temperature in wide band gap microcavities) [6]. Massive occupation of a polariton state has been recently observed in semiconductor two-dimensional (2D) cavities and attributed to Bose-Einstein condensation [7,8] or to polariton lasing [9]. Polariton condensation has also been claimed in a localized energy trap [10] with dimensions sufficiently large for the system to present a 2D continuum of states. In these experiments, the clear distinction of a thermodynamic phase transition (Bose-Einstein condensation) from a kinetic stimulated scattering (polariton lasing) is still debated.

In this Letter, we demonstrate polariton lasing in GaAs/GaAlAs micropillar cavities. In such zero-dimensional (0D) cavities, polariton states are confined in all directions and present a well-defined discretized energy spectrum [11,12]. The absence of translation invariance lifts the wave-vector conservation selection rules in polariton scatterings. In GaAs 2D microcavities, these selection rules are responsible for inefficient polariton-phonon or polariton-polariton scattering, preventing the buildup of a large occupancy in the lower energy states [13–16]. In this work, we show that polariton scattering is very efficient

in micropillars. Under nonresonant excitation, a threshold corresponding to a measured occupation factor equal to unity is observed, followed by a massive occupation of the lowest energy polariton state. At higher excitation power, the progressive transition from the strong to the weak coupling regime is evidenced with the onset of conventional photon lasing.

Changing the spatial excitation geometry, stimulation can be triggered on higher energy polariton states. Such behavior is characteristic of a polariton laser with competing stimulated scattering toward several polariton modes. It rules out Bose-Einstein condensation where only massive occupation of the ground state is expected. Finally the spectral blueshift of the polariton laser line is attributed to polariton self-interaction within the lasing mode. This experiment is the first demonstration of a solid-state matter-wave laser on 0D states.

Our sample, grown by molecular beam epitaxy, consists in a $\lambda/2$ $Ga_{0.05}Al_{0.95}As$ cavity surrounded by two $Ga_{0.05}Al_{0.95}As/Ga_{0.80}Al_{0.20}As$ Bragg mirrors with 26 and 30 pairs in the top and bottom mirrors, respectively. Three sets of four 7 nm GaAs QWs are inserted at the antinodes of the electromagnetic field: one set at the center of the cavity and the others at the first antinode in each mirror [17]. A wedge in the layer thickness allows continuous tuning of the cavity mode energy E_C with respect to the QW exciton energy (E_X), changing the detuning $\delta = E_C - E_X$. Square (of size 20–2 μm) and circular micropillars were fabricated along the wafer using electron beam lithography and reactive ion etching [see inset of Fig. 1(a)]. Photoluminescence (PL) experiments are performed on single micropillars using a cw Ti:sapphire laser focused onto a 3 μm diameter spot with a microscope objective. For excitation powers P exceeding 1 mW, the laser beam is chopped using an acousto-optic modulator with 1% duty cycle at 10 kHz. The emission is spectrally dispersed and detected with a nitrogen cooled CCD camera. The sample is maintained at 10 K in a cold finger cryostat. The laser is tuned to the first reflectivity minimum on the high energy side of the mirror stop-band around 1.68 eV.
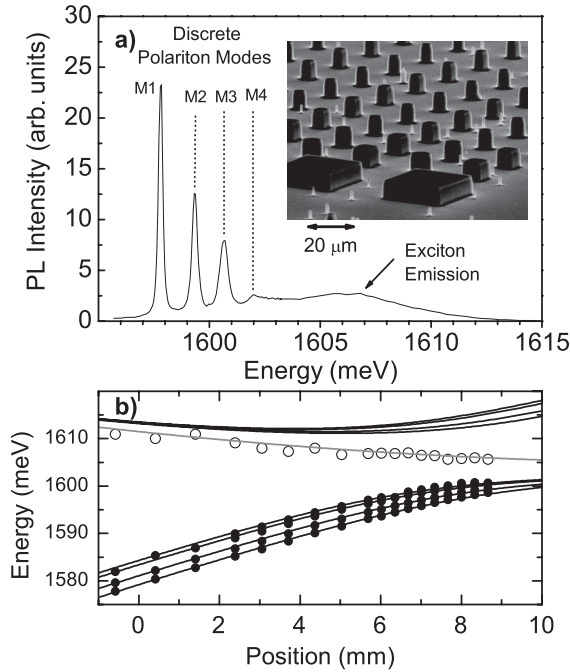
FIG. 1. (a) PL spectrum measured on a single 4 μm diameter micropillar; inset: scanning electron micrography of a micropillar array; (b) (black circles) emission energy of the discrete polariton PL lines and (open circles) of the exciton line measured on several 4 μm diameter micropillars along the cavity wedge, (thick black lines) calculated energy of the 0D polariton modes. $T = 10$ K.

In pillar cavities, photons are confined vertically by the Bragg mirrors and laterally by the index of refraction contrast between air and semiconductor. As a result, micropillars exhibit discrete 0D photon modes [11]. In the strong coupling regime, polaritons come from the mixing between each of these 0D photon modes and the QW excitons [12]. Figure 1(a) presents a PL spectrum measured on a single 4 μm diameter circular micropillar. The emission energies measured on micropillars of identical diameter along the cavity wedge are summarized in Fig. 1(b). The emission spectrum in Fig. 1(a) presents several discrete emission lines on the low energy side of the exciton line centered around 1607 meV. The energy of these discrete lines strongly varies with the layer thickness: they are attributed to 0D photon modes. For large negative detuning, the 130 μeV linewidth of these optical modes corresponds to a quality factor of 12 000. Each of these photon modes presents the characteristic anticrossing [18,19] with the exciton line. The 0D polariton energies are fitted using a 15 meV Rabi splitting. Note that contrary to 2D cavities, the uncoupled exciton line is observed because in-plane exciton emission is extracted through the pillar side. Since the exciton line is broadened by strain relaxation in the etching process, the upper polariton state could not be resolved.

PL spectra taken for increasing P on a 6 μm circular pillar are shown in Fig. 2. A spectrum taken at $P = 30$ μW

shows several discrete polariton modes. The polariton ground state corresponds to a 50% exciton 50% photon mixed state ($\delta = 0$ meV). Two excitation regimes can be distinguished. For $P > 5$ mW [Fig. 2(b)], the emission undergoes a pronounced blueshift and broadening. The density of electron-hole pairs per QW injected for $P = 5$ mW is estimated to be around 10^{10} cm^{-2} per QW, reaching the exciton screening density. In this excitation range, the strong coupling regime is progressively screened [20–22] and eventually the system enters the weak coupling regime, with emission from uncorrelated electron-hole pairs through the cavity modes. The strong coupling regime saturation is observed for excitation densities consistent with previous reports in 2D [20] or 0D [23] samples. Above 40 mW, a threshold is observed due to the onset of conventional photon lasing on the 0D photon modes [24].

Let us now concentrate on the strong coupling regime [$P < 5$ mW, Fig. 2(a)]. The emission behavior is marked by a sharp nonlinear increase of the ground-state PL intensity. As summarized in Fig. 3(a), when P varies from 0.3 mW up to 3 mW, the integrated intensity increases by 4 orders of magnitude. The occupancy N of the lowest energy polariton state can be experimentally estimated using: $N = I_{\rm PL}\tau_{\rm cav}/[\alpha^2 E]$, where $\tau_{\rm cav} = 3$ ps is the cavity photon lifetime, $I_{\rm PL}$ is the power emitted by the considered polariton mode, E its emission energy, and α its exciton part [13]. Relating the emission intensity to an actual polariton population may not always be possible since emission at the polariton energy could also come from correlated electron-hole pairs, as reported for bare excitons [25]. Nevertheless this analysis is valid at low density and
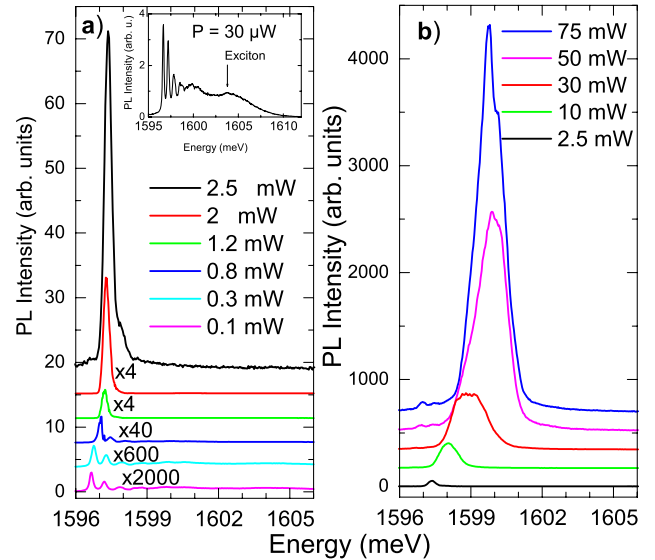

FIG. 2 (color online). Emission spectra measured for several excitation powers in a single 6 μm diameter micropillar. (a) Corresponds to the polariton lasing regime; (inset) emission spectrum measured for $P = 30$ μW. (b) Corresponds to the loss of the strong coupling regime and the onset of photon lasing. $T = 10$ K.
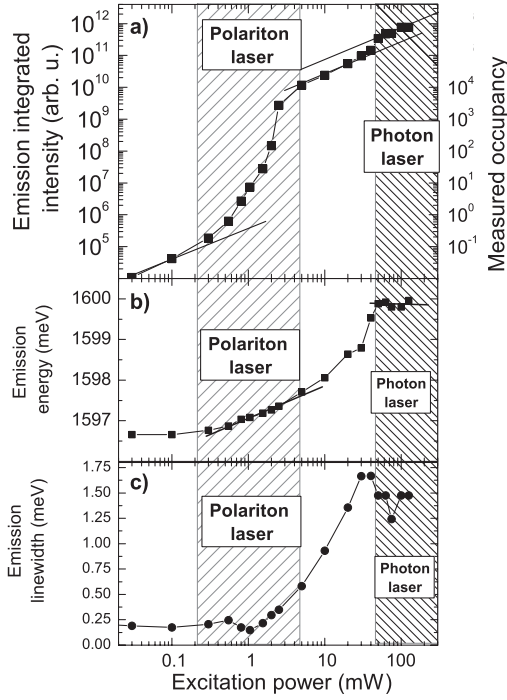
FIG. 3. (a) Integrated intensity and measured occupancy, (b) emission energy and (c) emission linewidth measured on the lowest energy emission line as a function of the excitation power; dashed areas highlight the excitation range for polariton lasing or photon lasing.
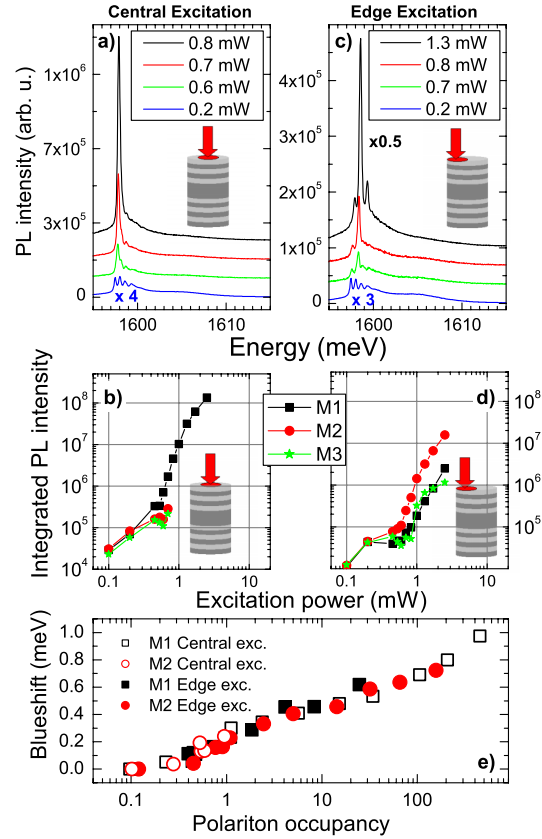

FIG. 4 (color online). (a) Emission spectra measured on a single 6 μm diameter micropillar for several excitation powers with a laser spot centered on the pillar surface, (b) measured integrated intensity of the three lowest energy polariton mode as a function of the excitation power in this centered excitation geometry; (c) and (d) same as (a) and (b) but with an edge excitation geometry as schematically indicated in the figure; (e) spectral blueshift of the polariton lasing modes as a function of their occupancy. $T = 10$ K.

low temperature, the regime where polariton nonlinearities are observed in the present Letter. As shown in Fig. 3(a), the onset of the polaritonic nonlinearity occurs when the measured polariton occupancy exceeds unity. Above threshold, the scattering of excitons toward the lowest energy polariton state is stimulated, leading to the formation of a macroscopically occupied polariton state. The polariton population of the lower energy state increases up to 10^4.

The polariton linewidth amounts to 0.2 meV at low excitation power, and slightly broadens with the onset of polariton-polariton interaction. At threshold, a spectral narrowing, down to ~0.15 meV, shows that the coherence time becomes longer than the radiative lifetime of single polaritons.

Polariton stimulated scattering is obtained on micropillars with diameter down to 2 μm and for detunings down to $\delta = -20$ meV. Working at higher temperatures, polariton lasing occurs up to 45 K. Above 50 K or for very large negative detunings, only the second nonlinearity associated to photon lasing is observed. Let us underline that in the unpatterned 2D sample, only conventional lasing was achieved [16]. Reducing the cavity dimensionality is the key step to achieve polariton quantum degeneracy under nonresonant excitation.

Figure 4 presents PL measurements on a 6 μm micropillar (with $\delta = -0.3$ meV for the lowest polariton mode) using two different excitation geometries. On the left part

of Fig. 4, the laser spot is centered on the micropillar surface. As described above, stimulated scattering occurs toward the lowest energy mode (M1). The data of the right part are recorded with the excitation spot shifted toward the micropillar edge. Under this excitation geometry, stimulated scattering is observed toward the first excited polariton states (M2). Further increasing the excitation power, competition between polariton modes also triggers stimulated scattering toward M1 and M3. This experiment demonstrates that the polaritonic nonlinearity cannot be interpreted in terms of a thermodynamic phase transition (as in Bose-Einstein condensation) because in this framework, the largest occupancy is always expected on the system ground state. In the present experiment, the nonlinearities are not governed by thermodynamics but by the kinetics of the scattering process toward the low energy polariton states. Such bosonic stimulation of polariton scattering has been named "polariton laser" [26,27] in analogy to the atom laser [28]. Nonetheless one must keep in mind that the stimulation mechanism is very differ-

ent from that in a conventional photon laser: it is not the emission of radiation that is amplified but a scattering mechanism, following the nonresonant excitation. As in a conventional photon laser, multimode polariton lasing can be triggered depending on the excitation condition. The electromagnetic field of the lowest energy mode (HE_{11}, see Ref. [12]) presents an antinode at the micropillar center and decays at the edge. It is therefore favored when the center of the pillar is excited. On the contrary, the second polariton line gathering three degenerate modes within the mode linewidth (EH_{01}, HE_{21}, and HE_{01}), is favored under edge excitation since these modes present an antinode at the periphery. Thus the pump excitation geometry triggers stimulated scattering toward polariton modes with matching field spatial distribution.

Contrary to conventional photon lasers, the lasing mode of a polariton laser is macroscopically occupied with half-matter half-light bosons, i.e., with interacting bosons. Exciton-exciton interaction and the resulting exciton blueshift has been extensively studied in the 1980s [29,30] and recently revisited within the framework of excitonic polaritons [31]. Polariton-polariton interaction originates from Coulomb interaction between the fermionic constituents (electron and hole) of their exciton part. As the occupancy of the polariton states increases, self-interaction within the lasing mode induces a spectral blueshift of the emission [32]. Figure 4(e) summarizes the spectral blueshift of M1 and M2 both under central and edge excitation conditions. The blueshifts are plotted as a function of the polariton occupancy, obtained by normalizing the emission intensity by the intensity at threshold. M3 is not reported because the contribution from HE_{12}, at higher energy than EH_{11} and HE_{41} [12] could not be correctly deconvoluted. The curves in Fig. 4(e) are strikingly identical regardless of the mode number or the excitation conditions. This indicates that the blueshift only depends on the number of polaritons within the considered state. For instance, when multimode lasing is achieved, each lasing mode presents its own blueshift, corresponding to its own occupancy. Thus the blueshift does not come from interaction with high energy excitons or electron-hole pairs, but mainly from the self-interaction energy within the considered mode. Notice that the present results evidence a blueshift logarithmically varying with the occupancy whereas theoretical calculations predicts a linear behavior [32]. Further theoretical investigation is probably needed to quantitatively describe the observed self-interaction energy.

To conclude, polariton lasing is demonstrated on the discrete modes of a GaAs/GaAlAs micropillar cavity. A sharp threshold associated with a spectral narrowing shows the onset of stimulated scattering toward the lowest energy polariton state. The polariton state occupancy is measured to reach 10^4. Changing the excitation spatial symmetry, multimode polariton lasing is triggered, demonstrating that the observed feature cannot be described in terms of Bose-Einstein condensation. Contrary to photon lasing obtained at higher excitation power, the polariton self-interaction within the macroscopically occupied state induces a continuous blueshift of the emission as the state occupancy builds up. These results, obtained in the well controlled GaAs semiconductor system, open the way toward an electrically pumped polariton laser [33] and will stimulate future experiments to investigate the emission quantum statistics of such a solid-state matter-wave laser.

*jacqueline.bloch@lpn.cnrs.fr
[1] A. Einstein, Sitzungsber. Preuss. Akad. Wiss. **1**, 3 (1925).
[2] M. N. Anderson *et al.*, Science **269**, 198 (1995).
[3] K. B. Davis *et al.*, Phys. Rev. Lett. **75**, 3969 (1995).
[4] A. Kavokin and G. Malpuech, *Cavity Polaritons* (Elsevier, Amsterdam, 2003).
[5] J. Keeling *et al.*, Phys. Rev. Lett. **93**, 226403 (2004); M. H. Szymanska, J. Keeling, and P. B. Littlewood, *ibid.* **96**, 230602 (2006).
[6] G. Malpuech *et al.*, Appl. Phys. Lett. **81**, 412 (2002).
[7] H. Deng *et al.*, Science **298**, 199 (2002).
[8] J. Kasprzak *et al.*, Nature (London) **443**, 409 (2006).
[9] S. Christopoulos *et al.*, Phys. Rev. Lett. **98**, 126405 (2007).
[10] R. Balili *et al.*, Science **316**, 1007 (2007).
[11] J. M. Gérard *et al.*, Appl. Phys. Lett. **69**, 449 (1996).
[12] G. Panzarini and L. C. Andreani, Phys. Rev. B **60**, 16799 (1999).
[13] P. Senellart *et al.*, Phys. Rev. B **62**, R16263 (2000).
[14] I. Tartakovskii *et al.*, Phys. Rev. B **62**, R2283 (2000).
[15] R. Butté *et al.*, Phys. Rev. B **65**, 205310 (2002).
[16] D. Bajoni *et al.*, Phys. Rev. B **76**, 201305(R) (2007).
[17] J. Bloch *et al.*, Appl. Phys. Lett. **73**, 1694 (1998).
[18] J. Bloch *et al.*, Superlattices Microstruct. **22**, 371 (1997).
[19] T. Gutbrod *et al.*, Phys. Rev. B **57**, 9950 (1998).
[20] R. Houdré *et al.*, Phys. Rev. B **52**, 7810 (1995).
[21] M. Kira *et al.*, Phys. Rev. Lett. **79**, 5170 (1997).
[22] H. Cao *et al.*, Phys. Rev. A **55**, 4632 (1997).
[23] E. S. Lee *et al.*, J. Appl. Phys. **89**, 807 (2001).
[24] Photon lasing is observed 5 meV below the bare cavity modes deduced from low density measurements. This effect, pronounced in cavities containing many QWs, can be understood considering changes of the QW layer refractive index when screening the exciton [16].
[25] S. Chatterjee *et al.*, Phys. Rev. Lett. **92**, 067402 (2004).
[26] A. Imamoglu and R. J. Ram, Phys. Lett. A **214**, 193 (1996); A. Imamoglu *et al.*, Phys. Rev. A **53**, 4250 (1996).
[27] I. A. Shelykh *et al.*, Phys. Rev. B **68**, 085311 (2003); F. P. Laussy *et al.*, Phys. Rev. Lett. **93**, 016402 (2004).
[28] H. M. Wiseman and M. J. Collett, Phys. Lett. A **202**, 246 (1995); M. Holland *et al.*, Phys. Rev. A **54**, R1757 (1996).
[29] N. Peyghambarian *et al.*, Phys. Rev. Lett. **53**, 2433 (1984).
[30] S. Schmitt-Rink, C. Ell, and H. Haug, Phys. Rev. B **33**, 1183 (1986).
[31] C. Ciuti *et al.*, Phys. Rev. B **62**, R4825 (2000).
[32] A. Verger, C. Ciuti, and I. Carusotto, Phys. Rev. B **73**, 193306 (2006).
[33] D. Bajoni *et al.*, Appl. Phys. Lett. **90**, 121114 (2007).