Microcavity polariton depopulation as evidence for stimulated scattering

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We report on a c.w. two-beam experiment unambiguously evidencing the polariton transfer from initial states to final states which takes place through a stimulated scattering process. One beam is nonresonant while the other resonantly creates a large occupation factor in the lower polariton final state. Gain, resulting in a deepening of the lower polariton reflectivity dip, is observed on the resonant beam. Simultaneously, the luminescence of large in-plane wave-vector states decreases linearly with the resonant power.

Since the observation of the strong-coupling regime (SCR) between excitons and photons in semiconductor microcavities (MC's),[1] many works have been performed[2] to understand the optical properties of such structures. The hope of using the SCR to modify the spontaneous emission dynamic has rapidly been weakened by the evidence of a long relaxation time encountered under nonresonant excitation. Indeed, in MC's, one exciton is coupled to one photon for a given in-plane wave vector k so that mixed exciton-photon states (polaritons) are formed. This one-to-one coupling deeply modifies the dispersion relation[3] so that the relaxation from large in-plane wave-vector excitons toward $k \approx 0$ states is very slow (bottleneck effect[4]). However, several recent works have reported on nonlinear emission[5–8] in the SCR indicating a collapse of this bottleneck.[5–7] The questions raised by these observations is whether they are a manifestation of the quasibosonic nature of polaritons. Indeed, for very low carrier densities, excitons are composite quasibosons. Thus, occupation factors close to one of the $k \approx 0$ states could trigger stimulated relaxation and superlinear emission.[9,10]

Nonlinear emission under nonresonant excitation has been observed both in II-VI and III-V MC's.[5,6] However, no definite evidence for the stimulated process appears from these one beam experiments. Recently, two beam experiments have been performed to evidence the stimulation process by resonantly creating a large final-state occupation. Savvidis *et al.* have taken advantage of the polariton dispersion to selectively excite polaritons for which the polariton-polariton scattering toward $k \approx 0$ states is very efficient.[11] They evidence a large gain on a probe beam at $k=0$ attributed to parametric amplification.[12] Parallely, Huang *et al.*[13] have observed an increase of the upper polariton emission when resonantly exciting the lower polariton (LP) both at $k=0$ and at larger k. As opposed to Ref. 11, this nonlinear effect is incoherent and lasts for ≈ 100 ps. These studies evidence a stimulated scattering looking at the final-state emission. However, to our knowledge, no evidence of the polariton transfer from initial to final states has ever been reported.

In the present paper, we unambiguously evidence the stimulated polariton transfer by performing a c.w. two-beam experiment. The first excitation is nonresonant. Angle-resolved measurements of the resulting emission show a relaxation bottleneck for low densities which collapses when increasing the nonresonant power. This collapse is accompanied by a nonlinear emission at $k \approx 0$. However, we show that this nonlinearity starts for very low occupation factors. To trigger stimulation, a resonant second beam directly creates a significant occupation factor in LP states at $k \approx 0$. We observe gain on the reflected resonant beam resulting in a deepening of the LP reflectivity dip. Parallely, the emission of large k states decreases as the resonant power increases. The simultaneous observation of amplification of the resonant beam and depopulation of large k states is the direct signature of the stimulated transfer. The balance between lost and gained signals is discussed as well as their dependence on excitation power, detuning, and temperature.

The experiment is performed on a III-V MC containing one $In_{0.05}Ga_{0.95}As$ 8 nm quantum-well and exhibiting a Rabi splitting of 3.5 meV. The sample is cooled at 2 K or 77 K using an immersion cryostat. The detuning ($\delta = E_{\mathrm{cavity}} - E_{\mathrm{exciton}}$) between the exciton and the photon modes is chosen by changing the position on the sample. Two c.w. Ti:sapphire laser beams are coupled to optical fibers having a 200 μm core. Both fibers are imaged on the sample in the same 100 μm diameter spot. One of the laser beams is tuned to the energy $E_{nr} = 1.61$ eV, beyond the mirror stop band. The other one, of energy E_r, is tuned across the LP resonance close to $k=0$. The excitation power of the nonresonant (resonant) beam is P_{nr} (P_r). Either beam can be chopped at low frequency ($\omega = 510$ Hz). To reduce broadening due epitaxial layer variations, we collect the emission of a 40 μm diameter spot within the excited spot. Angular selection is achieved by placing a diaphragm or a rectangular aperture after the collection lense. The emission is then coupled to a third optical fiber, spectrally analyzed by a spectrometer and detected by a Si multichannel detector, or a photomultiplier followed by either a photon counter or a lock-in amplifier.

We first analyze the angle-resolved photoluminescence (PL) emission with the nonresonant beam only. Figures 1(a) and 1(b) show PL spectra for two excitation powers P_{nr} and various external angles θ between 0° and 20° corresponding to k from 0 to 3×10^4 cm^{-1} [correspondence between k and θ can be seen in Fig. 1(f)]. Because of the strong dispersion,[3] angle-resolved PL selects emission from polaritons of different k. For low P_{nr}, the emission is larger around $k \approx 2$

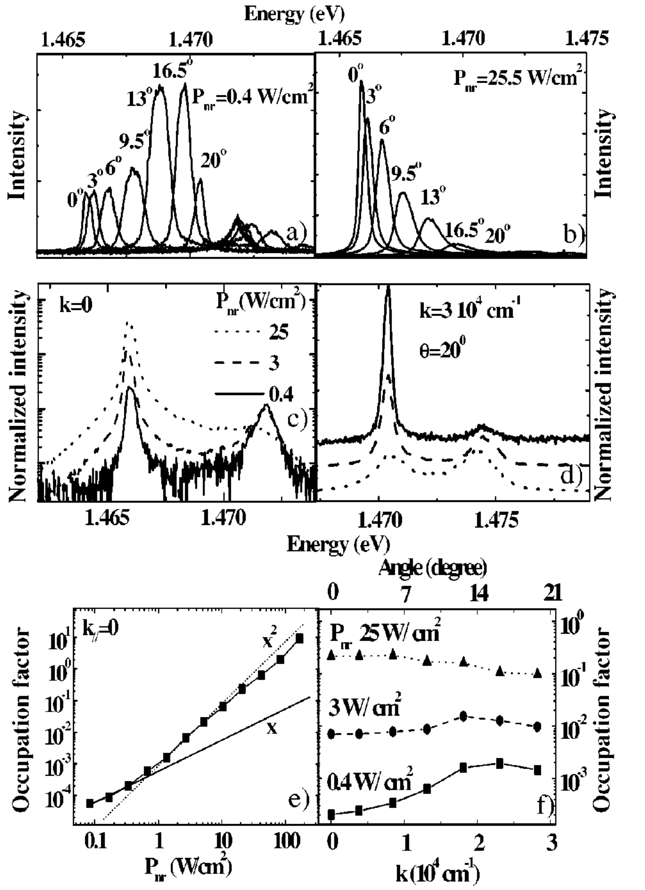

FIG. 1. $\delta=-5$ meV, $T=2$ K. (a),(b) PL spectra for various angles for two P_{nr}. (c),(d) PL spectra normalized to P_{nr} for three P_{nr}; (c) $\theta=0°$ (log. scale). (d) $\theta=20°$ (lin. scale). The spectra of (d) have been vertically shifted for clarity. (e) Occupation factor for $k=0$ as a function of P_{nr}. The straight (dotted) line shows what would be a linear (quadratic) dependence. (f) Occupation factor as a function of k or θ for three P_{nr}.

$\times 10^4$ cm^{-1} than for $k=0$ showing the relaxation bottleneck.[4,14] For higher P_{nr}, this bottleneck collapses and the emission is maximal around $k=0$. Figures 1(c) and 1(d) show PL spectra normalized to the incident power for $\theta=0$ and $\theta=20°$ and various P_{nr}. As we previously reported, the LP emission of $k\approx 0$ states is strongly superlinear.[6] Meanwhile, the integrated emission from large k LP states is linear and the lines significantly broaden. This broadening indicates that collisions become efficient and contribute to the bottleneck collapse.[14]

To find out whether the nonlinearity we observe is due to collisions only or also stimulated relaxation, we have estimated the occupation factors $f(\theta)$ of the emitting states from PL measurements.[15] To do so, we have calibrated our experimental setup to convert detector units into watt and relate the number of polaritons $N(\theta)$ to the PL emission in watt $I_{PL}(\theta)$: $N(\theta)=I_{PL}(\theta)*\tau_{cav}/[\alpha^2_{phot}(\theta)*E_{LP}(\theta)]$, where $\tau_{cav}=6$ ps is the cavity photon escape time, $\alpha_{phot}(\theta)$ is the LP photon weight, and $E_{LP}(\theta)$ its energy. The occupation factor is then given by $f(\theta)=N(\theta)/n_{\mathrm{states}}$, where $n_{\mathrm{states}}=(2\pi\delta k^2)*S/4\pi^2$ (S is the spot surface) is the number of states in the angular aperture $\delta k=2\times 10^3$ cm^{-1}. Notice that the PL signal is mainly due to a polariton population since,

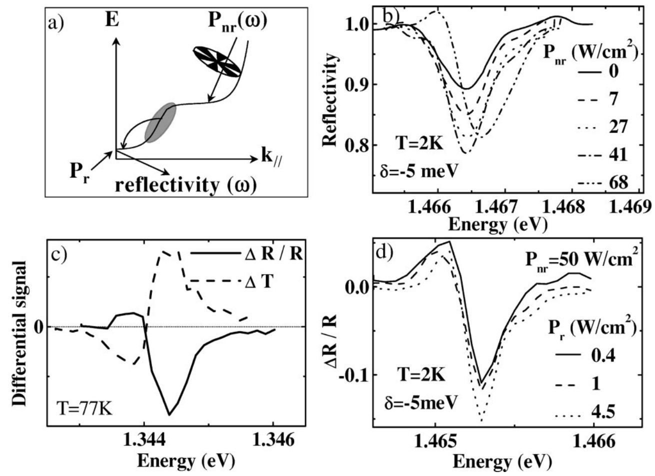

FIG. 2. (a) Schematic setup for differential reflectivity measurements. (b) Reflectivity spectra for various P_{nr}, $P_r=0.2$ W/cm^2. (c) Simultaneous measurement of the relative reflectivity change and differential transmission. (d) Relative reflectivity change for $P_{nr}=50$ W/cm^2 and various P_r.

as checked with time-resolved measurements,[16] the contribution of the instantaneous part, attributed to other mechanisms,[17] is negligible. Figure 1(e) shows the estimated occupation factor for $k=0$ as a function of P_{nr}. The nonlinearity begins when the occupation factor is below 10^{-3} showing that it is *not* due to stimulated scattering, contrary to our previous interpretation.[6] The quadratic increase of the occupation factor confirms the role of binary collisions in the bottleneck collapses. Occupation factors close to 1 are reached around $P_{nr}=40$ W/cm^2. Yet no second threshold is observed, maybe because the system is not far from the SCR bleaching limit (≈ 100 W/cm^2), or because our calibration procedure overestimates the occupation factors. Figure 1(f) shows how the occupation factor for the different k changes with excitation power.

With the nonresonant beam only, the range of powers for which occupation factors are close to 1 while the system is still in the SCR is narrow. Thus, it is difficult to evidence a stimulated process. In a second type of experiment, we turn on the resonant beam to directly create large occupation factors in $k\approx 0$ states. We first chop the nonresonant beam and analyze the intensity of the reflected resonant beam for various P_{nr} [see Fig. 2(a)]. As shown in Fig. 2(b), when increasing P_{nr}, the reflectivity dip becomes significantly more pronounced. Notice that as long as $P_{nr}<60$ W/cm^2, no shift of the line occurs. This deepening is observed both at 77 and at 2 K and for various negative detunings. Parallely, we have performed reflectivity and transmission measurements showing that simultaneously to the reflectivity changes described above, the transmitted intensity increases [Fig. 2(c)].[18]

As in previous reports,[19,20] for high carrier density ($P_{nr}>60$ W/cm^2), the polariton reflectivity dip broadens, weakens and shifts. Here we focus on experimental features occurring for lower P_{nr}, in a regime where screening effects, induced in particular by free carriers, are negligible. Another difference between experiments of Refs. 19 and 20 and ours, is that the resonant beam here is not a weak probe. Actually $P_r\approx 0.2$ W/cm^2 creates a polarization corresponding to a LP occupation factor of $f\approx 5$. The deepening of the LP line is

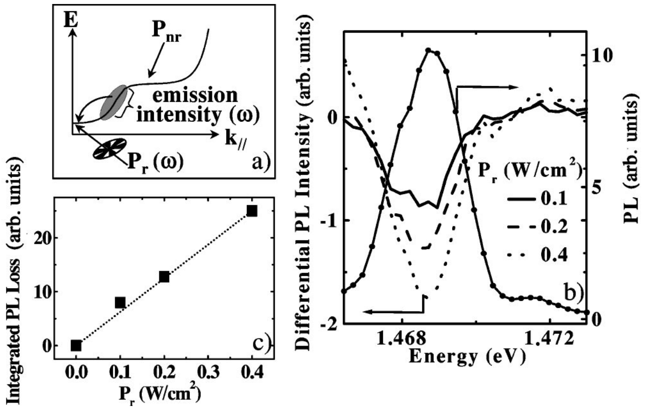

FIG. 3. (a) Schematic setup for differential PL measurements. (b) Right axis: PL spectrum measured between $\theta = 10°$ and 20° with $P_{nr} = 8\ \mathrm{W/cm^2}$ and $P_r = 0$. Left axis: Differential PL spectra between $\theta = 10°$ and 20° for $P_{nr} = 8\ \mathrm{W/cm^2}$ and various P_r. The same units are used for right and left axis. (c) Integrated PL loss as a function of P_r. The dotted line is a linear fit. $\delta = -5$ meV and $T = 2$ K.

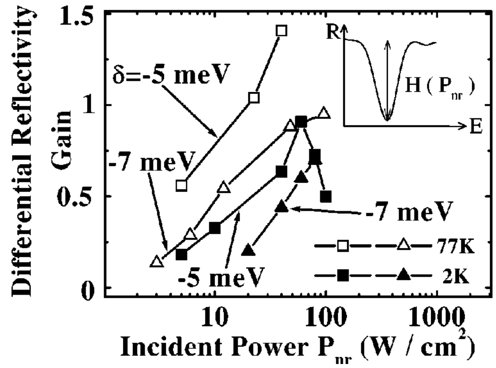

FIG. 4. Differential reflectivity gain (as defined in the text) as a function of P_{nr} for two temperatures and detunings.

the signature of a gain mechanism as can be understood qualitatively as follows. The reflected intensity is the result of a destructive interference between light reflected on the surface and light that has propagated inside the cavity. If the field gets larger inside the cavity, then the intensity of the reflected beam decreases (a result of more efficient destructive interference),[21] and the transmitted beam intensity increases. Two mechanisms could lead to an increase of the field inside the cavity: a reduction of the residual absorption or an amplification. Residual absorption is measured to be negligible and cannot explain the reflectivity change we observe.[22] Thus, the deepening of the LP reflectivity dip as well as the transmission changes are due to *gain* on the resonant beam.

Figure 2(d) shows the relative reflectivity change observed for a given P_{nr} and various P_r. The spectra are almost superimposed showing that the resonant beam never saturates the gain mechanism. Indeed, even though the resonant beam locally creates a large LP occupation, it creates a total number of carriers much smaller than the one injected by the nonresonant beam. Moreover, within the picture of stimulated relaxation process, the modification of the reflected beam intensity $\Delta I_r(\omega) = I_r^{P_{nr}} - I_r^{P_{nr}=0}$ is proportional to $(1+f)$. As $f \gg 1$ and is mainly due to the resonant beam: $\Delta I_r(\omega) \propto P_r$. Since the reflected signal with $P_{nr}=0$ also follows $I_r^{P_{nr}=0} \propto R P_r$, then $\Delta R(\omega)/R = \Delta I_r(\omega)/I_r^{P_{nr}=0}$ is independent of P_r in a first-order approximation.

Within the picture of a stimulated scattering, we expect the emission of large k states to decrease when $P_r \neq 0$. To check that hypothesis, we monitor the influence of the resonant beam on the emission of large k states. The chopper is now placed on the resonant beam as shown in Fig. 3(a) and P_{nr} is set to $8\ \mathrm{W/cm^2}$. We measure the PL emission from states between 10° and 20° using a rectangular aperture after the collection lense. Figure 3(b) shows that the PL spectrum obtained in the absence of the resonant beam peaks around ≈ 1.469 eV ($k \approx 2 \times 10^4\ \mathrm{cm^{-1}}$). When P_r is on, we measure the modulated component of the emission corresponding to PL variations induced by the resonant beam. This modulated signal is shown in Fig. 3(b) for various P_r. A *reduction* of the emission of large-k states is observed. For the same excitation values of P_r and P_{nr}, gain is evidenced in the reflectivity measurements [see Fig. 2(b)]. This simultaneous observation of gain at $k \approx 0$ and loss at larger k is a clear evidence for stimulated scattering.

Let us study the dependence of the PL diminution on P_r. In Fig. 3(b), a positive ΔPL signal is observed on the low-energy side of the spectrum for the highest P_r. Indeed, collision processes between carriers injected by the two beams are responsible for an increase of PL in a wide range of k, which appears as a background superimposed to the negative dip in the differential PL signal. In Fig. 3(c), we plot the area of this dip, disregarding the positive background, as a function of P_r. The PL loss varies linearly with P_r, a behavior consistent with the picture of a stimulated scattering rate proportional to the final-state population. This is of course true because the initial state is only weakly depleted: the maximum PL loss amounts to about 20% and we know from Fig. 2(d) that the resonant beam never saturates the gain.

Finally, we try to evaluate the number of polaritons transferred through the stimulated scattering. $P_r = 0.4\ \mathrm{W/cm^2}$ creates about 750 polaritons around $k \approx 0$. For $P_{nr} = 8\ \mathrm{W/cm^2}$, since the increase of the reflectivity dip is $\approx 30\%$, about 225 polaritons are transferred toward $k = 0$. Using again our setup calibration, we deduce that the reduction of the large k PL signal corresponds to a loss of about 100 polaritons (we assume an isotropic loss in $\vec{k}$ space). Since many large k exciton states not accessible in this experiment are also populated by the nonresonant beam and certainly contribute to the stimulated transfer toward $k \approx 0$, the apparent discrepancy between the number of transferred polaritons in PL and reflectivity is not surprising. Our estimation gives the right order of magnitude.

Finally, we report on additional experimental features. We define the differential gain observed in reflectivity as the dip increase

$$\frac{H(P_{nr}) - H(P_{nr}=0)}{H(P_{nr}=0)},$$

$[H(P_{nr})$ is defined in the inset of Fig. 4]. This gain is plotted for two detunings and temperatures in Fig. 4. For a given P_{nr}, the gain value for $\delta = -5$ meV is about twice as large as for $\delta = -7$ meV. This difference reflects the exciton polariton weight which is $\alpha_{exc}^2 = 0.09$ for $\delta = -5$ meV as opposed to 0.05 for $\delta = -7$ meV. Another interesting feature is the temperature dependence. For a given δ, a nonresonant power ≈ 5 times smaller is needed at 77 K than at 2 K to observe the same gain value. This strong variation on temperature confirms that the dominant scattering mechanism is collisions and not phonon scattering which is expected to vary slowly with temperature.[10]

Lastly, notice that the present experiment enables to evidence the stimulated scattering in an interesting density range. P_{nr} is typically 10 W/cm^2 and creates a carrier density of about 10^9 cm^{-2}. For such densities, the bosonic approximation for excitons is actually justified.[23] The resonant beam creates occupation factors of the order of 10 only locally in k space. The gain process observed here is quite different from Ref. 11 where a coherent amplification occurs within a few ps after the laser pulse only. Here as in Ref. 13, gain comes from the scattering from an incoherent polariton population (created in our case by a nonresonant second laser). In addition, the observation of reflectivity changes confirms that in this stimulated transfer, polaritons of large k do get the phase of the polaritons created by the resonant beam.

To conclude, the estimation of the occupation factors shows that nonlinear emission under a single nonresonant excitation is due to collisions and not to stimulated scattering. Our two-beam experiment evidences gain on the $k=0$ final states simultaneously to a decrease of large k initial-state population. Both gain and loss are linear with the number of polaritons injected in the final state. These measurements directly show the transfer from initial to final state through a stimulated scattering.

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