Electron-Polariton Scattering in Semiconductor Microcavities

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In semiconductor microcavities, electron-polariton scattering has been proposed as an efficient process that can drive polaritons from the bottleneck region to the ground state, achieving Bose amplification of the optical emission. We present clear experimental observation of this process in a structure that allows control of the electron density and we report substantial enhancement of photoluminescence. We show that this enhancement is more effective at higher temperatures due to the different way that electron scattering processes either broaden or relax polaritons.

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Polaritons are the half-exciton, half-photon quasiparticles resulting from the strong coupling between quantum well (QW) excitons and cavity photons in semiconductor microcavities [1,2]. These quasiparticles are very light and so their de Broglie wavelength is very large. Therefore quantum degeneracy of a polariton gas is achievable at much lower densities than for an exciton gas [3]. However, polaritons are composite bosons and the fermionic nature of electrons and holes appears as soon as the excitons spatially overlap [4]. A key point concerning the bosonic properties of polaritons has been therefore to establish whether the density necessary to reach quantum degeneracy is higher or lower than the density at which excitons ionize.

Recently, observations of parametric polariton scattering under stimulation of the final state gave conclusive proof of the bosonic behavior of polaritons [5] and brought to the fore the inherently contradictory views of Bose condensation phenomena in excitonic systems [3]. Among the many speculations on the advantages of polariton condensates is that of a new generation of optoelectronic devices based on a polariton laser or "*plaser*" [6–8]. However, Bose condensation of polaritons has not yet been observed from an initially incoherent population of excitons. The main obstacle has been a relaxation bottleneck which prevents excitons from rapidly relaxing into the strongly optically coupled polariton states at low energy [9–11]. Surprisingly, strongly coupled semiconductor microcavities (MCs) are found to be *poor* emitters at low carrier density because of this bottleneck. Polariton-polariton scattering has been found to be insufficiently effective at bypassing this bottleneck for carrier densities below the limit which retains strong coupling (SC) [12–15]. Recent theoretical studies propose an electron-polariton scattering relaxation mechanism as an excellent candidate for the Bose condensation of polaritons [16,17]. Similar mechanisms have been carefully considered for various cases of electron-exciton scattering in bulk semiconductors [18]. In planar MCs the modified energy dispersion around $k = 0$ allows these processes to be further disentangled experimentally.

In this paper we obtain clear experimental evidence for the assistance of electron-polariton scattering in breaking the bottleneck and opening the way towards condensation in excitonic systems. If a population of free electrons is introduced within the active region, polaritons can be scattered from the bottleneck region to the bottom of the lower polariton (LP) branch by giving their excess energy and momentum to free electrons (Fig. 1). The excited electrons are then rapidly cooled by the reservoir of cold electrons in the structure thus providing a "thermal lock" for the scattered polaritons at low temperatures [19]. Under these circumstances, polaritons at the bottom of the LP branch ($k \sim 0$) are effectively confined in a polariton trap (Fig. 1). The electrons facilitate rapid relaxation because (i) charge-dipole scattering is so much stronger than dipole-dipole scattering, and (ii) the electron is lighter than excitons and can take away more energy for a given momentum. Since the effectiveness of the process depends on the concentration of free

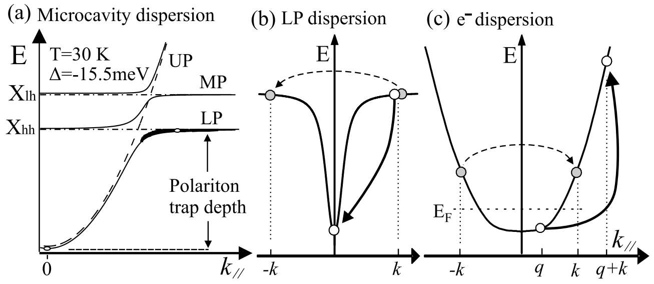

FIG. 1. (a) Dispersion relations of upper, middle, and lower polariton modes (solid lines), cavity (dashed line), X_{hh} and X_{lh} modes (dash-dotted lines) for $\Delta = -15.5$ meV and $T = 30$ K. (b),(c) Initial and final states of electron-polariton scattering that increases the polariton population at the bottom of the trap (solid arrow), and for elastic scattering (dashed arrow), plotted on both the LP branch (b) and the electron dispersion (c).

electrons in the active region, we use a phototunable n-doped heterostructure that allows for fine modulation of the free carrier density. We demonstrate that while electron-exciton scattering gets weaker as the temperature increases, the scattering to the low-energy polariton states actually gets stronger due to energy-momentum conservation.

The semiconductor MC structure used in this study was grown by molecular beam epitaxy on GaAs and incorporates 15 (25) periods of AlAs/AlGaAs distributed Bragg reflectors on the top (bottom). respectively [20]. In the center of the intracavity spacer is a mixed type-I/type-II QW structure (MTQW) consisting of a single 200 Å GaAs wide QW, clad on both sides by 26 Å GaAs narrow QWs separated by 102 Å AlAs barriers. The symmetry of the structure suppresses electric fields caused by the separation of e and h into different QWs. The optical cavity length is $\sim\lambda$ and varies across the sample. The SC of the cavity mode with both heavy and light hole excitons (X_{hh} and X_{lh}) in the GaAs wide QW results in three polariton branches (Fig. 1). When a two-dimensional electron gas (2DEG) is introduced into the structure a fourth excitonic mode appears due to the formation of trions [21]. In the current work, we do not explicitly extract the trion emission because it appears as a very weak line, disappearing rapidly as we raise the temperature, in contrast to the scattering observed here.

The structure employed for this study allows fine-tuning of the 2DEG in the wide QW within a single sample/device. Under laser photoexcitation at photon energies below the band gap of the narrow QW, excitons can be generated only in the wide QW. However for weak laser photoexcitation with photon energies above the narrow QW band gap, an electron-hole plasma is excited both in the wide QW and in the narrow QW. The MTQW architecture allows for efficient resonant transfer of electrons from the narrow QW to the wide QW through the X valley of the barrier layer, while holes are trapped in the narrow QW by the absence of these resonant states. This configuration provides an efficient way to repeatably vary the exciton and electron densities in the wide QW. In this study a continuous wave (CW) Ti:sapphire laser at 1.557 eV excites exclusively the wide QW and a diode laser at 1.952 eV varies the electron density, n_e. The Ti:sapphire laser is set to excite the cavity mode resonantly. Although the exciton density is accurately estimated from the intensities of the Ti:sapphire laser, I_X, and the diode laser, I_n, the exact dependence of n_e on the intensity of the diode laser is not known. The range of injected n_e used in this study is estimated to be $0 < n_e < 10^{12}\ \mathrm{cm}^{-2}$. Previous studies assumed a linear dependence $n_e \propto I_n$, however more recent calculations predict a nonlinear behavior [17,20]. Therefore instead of using n_e, in what follows we will express all dependencies as a function of the diode laser intensity.

Photoluminescence (PL) spectra are taken at normal incidence (i.e., in-plane $k = 0$), for $T = 5$ to 30 K, at low excitation powers of both wide and narrow QWs to preserve SC. The emitted light is collected in a cone of ±0.15°. Under these conditions the PL from the lower and middle polariton (MP) branches is modified by introducing a 2DEG in the wide QW. This change of PL varies from enhancement to suppression depending on the exact resonance of the cavity mode with the X_{hh} [Fig. 2(a)]. For this detuning dependence we fix a moderate n_e in the wide QW at a temperature of 30 K in order to avoid any effects from charged exciton states. For each illumination spot we normalize to the absorbed power of the wide QW, measure the PL enhancement $\Delta PL = [PL(I_X + I_n) - PL(I_n)]$ (with respect to the lowest power PL measured), and calculate $\eta = 100\% \times \Delta PL/[PL(I_X) + PL(I_n)]$. It is evident that for a wide range of negative detunings the presence of a 2DEG is beneficial to the PL. This implies that whenever the polariton trap is deeper than a few meV (and thus the relaxation bottleneck is present on the LP branch), electrons enhance the scattering from the bottleneck region to the ground state at $k = 0$ [Fig. 2(a)]. The detuning dependence of the PL enhancement follows the same trend independently of both temperature and CW excitation intensity, as long as the system remains in the SC regime. This independence arises because the shape of the LP branch remains virtually unaffected with increasing temperature.

In order to reveal the extent of this enhancement, we measure ΔPL as a function of the electron density for a range of different detunings. With the cavity mode optimally detuned from the X_{hh} ($\Delta = -15.5$ meV), the

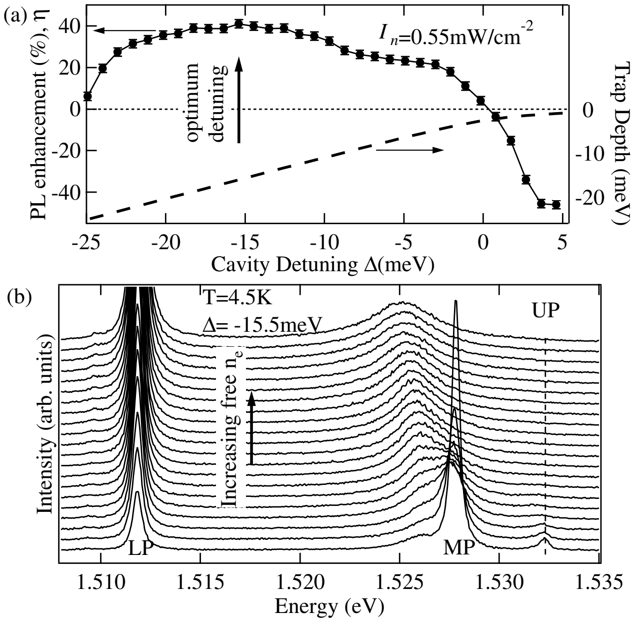

FIG. 2. (a) Photoluminescence enhancement of the lower polariton mode (solid line) and polariton trap depth (dashed line) as a function of the cavity detuning energy. (b) Photoluminescence spectra for different free electron densities n_e (n_e increases upwards).

normally emitted PL spectra are recorded in Fig. 2(b) as n_e increases (upwards). Here the weak Ti:sapphire pump, $I_X = 20\ \mathrm{mW\,cm^{-2}}$, excites only exciton densities $n_X \simeq 4 \times 10^9\ \mathrm{cm^{-2}}$ (assuming $\tau_X \sim 500$ ps). With free electrons injected into the wide QW, the polariton modes are drastically modified, as a result of electron-exciton scattering. The PL intensity of the LP mode increases rapidly [Fig. 3(a)], while the PL from the MP mode conversely decreases. However, increasing n_e also systematically broadens the MP mode and redshifts its peak energy position [Fig. 3(b)]. The sublinear enhancement follows $\sim I_n^{0.6}$ below a critical diode power, I_n^{sat} (regions I and II), above which the slope reduces to $\sim I_n^{0.4}$ (region III). To understand the nature of this threshold we note that the redshift of the MP mode energy towards the bare X_{hh} energy marks a different critical diode power, I_n^{WC}, at which the system passes from a strong coupling (region I) into the weak coupling (WC) regime (regions II and III). The dependence of the PL enhancement on I_n remains *virtually unaffected* by this transition to the WC regime. This is due to the negligible difference in properties of the LP branch between the SC and WC regimes at these negative detunings (Fig. 1). Even in the WC regime, the lower cavity mode is weakly coupled to the bare exciton and the bottleneck in relaxation remains. In structures with a small Rabi splitting (here only 4 meV), the transition to the WC regime occurs without substantially collision broadening the exciton transition, so that excitons remain as well-defined states. Electron-exciton scattering thus exhibits similar properties in both the SC and WC regimes.

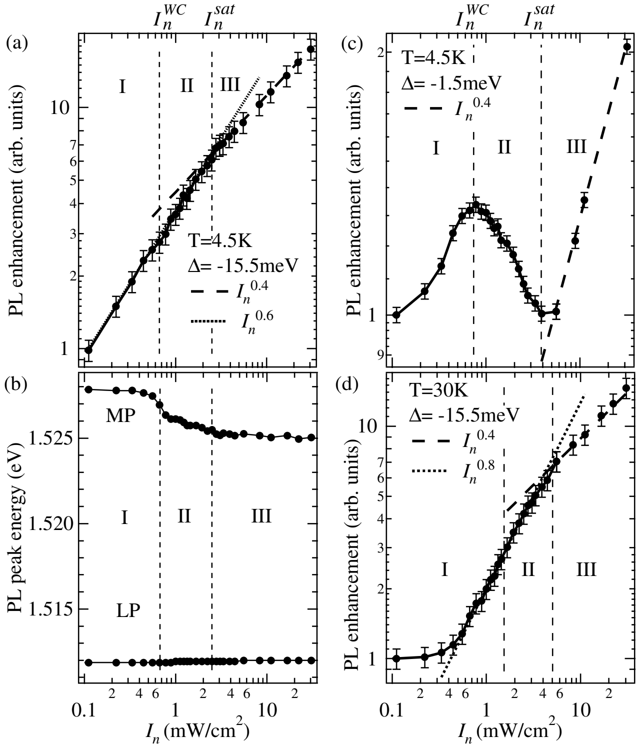

FIG. 3. (a) PL enhancement vs I_n for $\Delta = -15.5$ meV at $T = 4.5$ K. Vertical dotted lines separate the three different regions that correspond to the strong coupling regime (I), weak coupling regime (II), and ionized excitons (III). (b) Peak PL emission energies for the lower and middle polariton branches vs I_n. (c),(d) Same as (a) but for (c) $\Delta = -1.5$ meV at $T = 4.5$ K, and (d) $\Delta = -15.5$ meV at $T = 30$ K.

The rate of PL enhancement decreases only after the electron density becomes high enough (at I_n^{sat}) to dissociate the excitons in the wide QW, $n_e > 10^{11}\ \mathrm{cm^{-2}}$. To confirm that a separate contribution to the PL enhancement indeed exists when the excitons are ionized, measurements are shown for another detuning condition ($\Delta = -1.5$ meV) at which the SC and WC dispersion relations vary considerably, but for which a low n_e still enhances the LP mode emission [Fig. 2(a)]. The PL enhancement with I_n shown in Fig. 3(c) is again separated into three regions. The first vertical dashed line corresponds to the transition from the SC to the WC regime, as identified from the shift of the peak energy positions. For this small negative detuning the LP branch and the bare cavity mode differ greatly, unlike the situation at very negative detuning. Therefore, for a wide range of n_e (regions I and II) the excitons are still well-defined quasiparticles and electron-exciton scattering is a well-defined process. In this case when the system enters the WC regime ΔPL changes behavior because the LP and the bare cavity mode now have different shapes. However in region III, the enhancement in emission is once again sublinear with the same exponent as for the optimum detuning, $\Delta PL \propto I_n^{0.4}$. This confirms that region III indeed corresponds to the screening out of the bound exciton and that the recombination of the electron-hole plasma in the wide QW depends on the excess electron density, independent of detuning.

Although an enhancement in the PL at $k = 0$ is seen at $T = 4.5$ K, it is limited to a factor of 2, within the SC regime. One of the main reasons for this is that the extra homogeneous broadening of the exciton transition produced by injecting the 2DEG leads to a rapid reduction of the exciton oscillator strength and the transition to the WC regime. However we find this relationship between PL enhancement and exciton broadening produced by the electron-polariton scattering is considerably affected by temperature. The electron-induced PL enhancement at $T = 30$ K and the optimum resonance condition ($\Delta = -15.5$ meV) is shown in Fig. 3(d).

In this case the PL enhancement turns on only after a larger diode power and similarly, both the transition to the weak coupling, I_n^{WC}, and the exciton unbinding, I_n^{sat}, are shifted to a higher diode power (by a factor ~2.5). This would suggest that electron-polariton scattering is *reduced* at higher temperatures as predicted by recent theoretical calculations [17]. At the same time, in the strong coupling regime the enhancement of PL by electrons is more rapid, $\Delta PL \propto I_n^{0.8}$, implying that the electron-polariton scattering rate is *larger* at higher temperatures (discussed below). Within the SC regime this immediately results in a threefold enhancement of PL due to electron-polariton scattering compared to PL emitted without photoinjecting electrons into the wide QW. Once again in the plasma regime, the dependence on I_n remains unchanged with temperature for the reasons discussed above. However, before the excitons are ionized, a tenfold enhancement is achieved.

The smaller contribution to exciton broadening by electrons at higher temperatures is confirmed by plotting the linewidth of the (mostly X_{hh}-like) MP mode vs temperature at low n_e (Fig. 4). This observation agrees with calculations of the electron-exciton interaction matrix elements, which favor small energy transfer transitions, as seen in Fig. 1 on both electron and polariton dispersions with dashed arrows [17]. Essentially, as the exciton temperature increases, energy-momentum conservation reduces the range of final states that electrons can scatter excitons into and decreases the strength of this scattering. Consequently, the higher the temperature, the less effective electrons will be in the electron-exciton scattering interaction. This would indeed mean that a larger electron density is needed both to reduce the exciton oscillator strength enough to pass over to weak coupling, and also to ionize the excitons, in agreement with the increase in both I_n^{WC} and I_n^{sat} at $T = 30$ K. The linewidth narrowing of the MP mode with increasing temperature (Fig. 4), in contrast with the linewidth broadening of the MP mode with increase in CW excitation intensity [Fig. 2(b)], shows that the CW excitation intensity does not act through increasing the width of the exciton levels due to sample heating.

To understand how the efficiency of electron-polariton scattering (observed in enhanced PL) *increases* despite the drop in electron-exciton scattering at higher temperatures, we need to understand the very different scattering processes participating. As depicted in Fig. 1 (solid arrows), the scattering of an exciton from the bottleneck region to $k \approx 0$ implies a small exchange in wave vector but a large exchange of energy. This exchange favors electrons with initial states of higher energy/wave vector as found at higher temperatures. This effect is thus not in contradiction with the previous analysis since it implies that only the specific scattering rate (into the $k_{\mathrm{final}} \approx 0$) becomes faster while the scattering contribution to the exciton linewidth involves all final states. The approximation of a specifically thermal electron distribution is of minor importance in the simplified picture of the proposed relaxation mechanism. However further experiments and theory should clarify the actual electron distribution.

A number of approximations are made in this model. Our theory neglects the effects of bound charged exciton states; however recent calculations show that trion dissociation operates in a similar manner to the free particle scattering presented here [17]. Trion-mediated scattering will therefore contribute in the same way as electrons at low temperatures, but should be absent above $T = 10$ K due to their ionization [22]. In addition, we ignore the effects of both exciton and carrier localization, because the samples are of high quality with polariton linewidths 8 times smaller than the Rabi splitting.

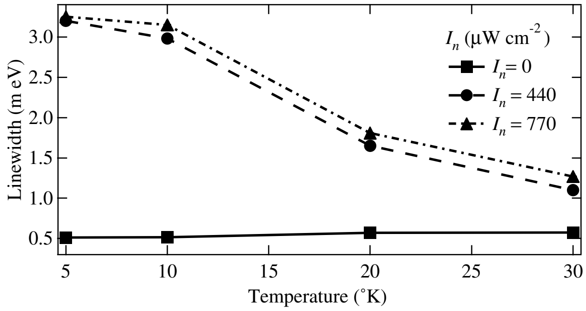

FIG. 4. Linewidth (FWHM) of middle polariton mode as a function of temperature at several I_n.

This paper thus presents compelling evidence for the effectiveness of electrons within the active region of a strongly coupled semiconductor MC, for enhancing relaxation of polaritons. This matches recent theoretical predictions. Understanding the ways that electron-exciton Coulomb scattering contributes differently to exciton broadening and PL enhancement paves the way to improved polariton emitter and lasers.

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[1] C. Weisbuch *et al.*, Phys. Rev. Lett. **69**, 3314 (1992).
[2] M. S. Skolnick, T. A. Fisher, and D. M. Whittaker, Semicond. Sci. Technol. **13**, 645 (1998).
[3] L. V. Keldysh, *in Bose-Einstein Condensation*, edited by A. Griffin, D. W. Snoke, and A. Stringari (Cambridge University Press, Cambridge, England, 1995).
[4] J. Fernandez-Rossier *et al.*, Phys. Rev. Lett. **78**, 4809 (1997).
[5] P. G. Savvidis *et al.*, Phys. Rev. Lett. **84**, 1547 (2000).
[6] G. Malpuech *et al.*, Appl. Phys. Lett. **81**, 412 (2002).
[7] P. Senellart *et al.*, Phys. Rev. Lett. **82**, 1233 (1999).
[8] R. M. Stevenson *et al.*, Phys. Rev. Lett. **85**, 3680 (2000).
[9] M. Müller *et al.*, Phys. Rev. B **62**, 16 886 (2000).
[10] A. I. Tartakovskii *et al.*, Phys. Rev. B **62**, 2283 (2000).
[11] F. Tassone *et al.*, Phys. Rev. B **59**, 10 830 (1999).
[12] Le Si Dang *et al.*, Phys. Rev. Lett. **81**, 3920 (1998).
[13] R. Butté *et al.*, Phys. Rev. B **65**, 205310 (2002).
[14] R. Houdré *et al.*, Phys. Rev. B **52**, 7810 (1995).
[15] B. Sermage *et al.*, Phys. Rev. B **53**, 16 516 (1996).
[16] G. Malpuech *et al.*, Phys. Rev. B **65**, 153310 (2002).
[17] G. Ramon *et al.*, Phys. Rev. B **67**, 045323 (2003).
[18] C. Klingshirn *et al.*, Phys. Rep. **70**, 315 (1981).
[19] P. G. Savvidis *et al.*, Phys. Rev. B **65**, 073309 (2002).
[20] R. Rapaport *et al.*, Phys. Rev. B **63**, 235310 (2001).
[21] R. Rapaport *et al.*, Phys. Rev. Lett. **84**, 1607 (2000).
[22] A. Qarry *et al.*, Phys. Rev. B **67**, 115320 (2003).