Electron-polariton scattering, beneficial and detrimental effects

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We study the effects of electron-polariton scattering in a semiconductor microcavity that allows control of the electron density. We show that this process can efficiently drive polaritons to the ground polariton state and that it is more effective whenever the relaxation bottleneck is present on the LP branch.

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1 Introduction

Strong interaction between cavity photons and quantum well excitons results in new eigenstates of the system, the polaritons [1, 2]. Polaritons exhibit a very steep dispersion around the ground polariton state resulting in a much longer polariton de Broglie wavelength compared to that of bare excitons. 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. Due to the effect known as relaxation bottleneck, which prevents polaritons from relaxing into their ground state, Bose condensation of polaritons has not yet been observed from an initially incoherent population of excitons.

Recently, observations of parametric polariton scattering under stimulation of the final state gave conclusive proof of the bosonic behaviour 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 to rapidly relax 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].

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Fig. 1 (a) Dispersion relations of upper, middle and lower polariton modes (*solid lines*), cavity, X_{hh} and X_{lh} modes (*dashed 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).

Recent theoretical studies propose an electron-polariton scattering relaxation mechanism as an excellent candidate for driving polaritons to Bose condensation phenomena [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. 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, as seen by solid arrows in figure 1b, c. 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, as they lose the excess of energy needed to ionize the ground state polaritons [19]. Under these circumstances, polaritons at the bottom of the LP branch (k \approx 0) are effectively confined in the polariton trap. The electrons facilitate rapid relaxation of polaritons because (a) charge-dipole scattering is much stronger than dipole-dipole scattering, and (b) electrons are lighter than excitons and can take away more energy for a given momentum.

In this paper we review our recent experimental observation for the effectiveness of electron polariton scattering in breaking the bottleneck [20]. Since the effectiveness of the process depends on the concentration of free electrons in the active region, we use a photo-tunable *n*-doped heterostructure that allows for fine modulation of the free carrier density. We demonstrate that for very negative detuning, the effects of electron polariton scattering remain unaffected by the transition to the WC regime due to the negligible difference in the shape of the LP branch between the SC and WC regime.

2 Experimental configuration

The semiconductor microcavity structure used in this study was grown by molecular beam epitaxy on GaAs and incorporates 15 (25) periods of AlAs/AlGaAs distributed Bragg reflectors (DBR) on the top (bottom) [21]. 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 symmetry of the structure suppresses electrons and holes into different quantum wells (QWs). The strong coupling (SC) of the cavity mode with both heavy and light hole excitons (X_{thh} and X_{th}) in the GaAs wide-QW results in three polariton branches (Fig. 1a). When a two-dimensional electron gas (2DEG) is introduced into the structure, a fourth excitonic mode appears due to the formation of trions [22, 23]. In the current study, the trion emission has not been explicitly extracted because it appears as a very weak line, disappearing rapidly with rising temperature, in contrast to the scattering observed here.



Fig. 2 Photoluminescence enhancement of the lower polariton mode (*solid line*) and polariton trap depth (*dashed line*) as a function of the cavity detuning energy.

The mixed type-I/type-II QW (MTQW) structure was employed for this investigation since it allows for 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 only be generated 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 Xvalley 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 exclusively excites the wide QW and a diode laser at 1.952 eV varies the electron density, ne. The Ti:Sapphire laser is set to excite the cavity mode quasi-resonantly, i.e. at high angles and energies above the polariton region. 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 $0 < n_e < 10^{12} \text{ cm}^{-2}$ according to previous studies that assumed a linear dependence $n^{e} \propto I_{n}$, although more recent calculations predict a non-linear behaviour [17, 21]. Therefore, instead of using n_e , all dependencies are expressed as a function of the diode laser intensity.

3 Resonance dependence

The effectiveness of electron polariton scattering on the polariton energy transfer between the bottleneck region and the ground polariton state was investigated by changing the energy resonance between the bare cavity mode and the bare exciton mode. In this way we smoothly tune the energy of the ground polariton state, while the energy of the bottleneck region remains unaffected, and therefore we change the amount of energy a polariton needs to exchange in order to scatter from the bottleneck region to the ground polariton state. Photoluminescence (PL) spectra were taken at normal incidence (i.e. in-plane k = 0), for T = 5 K to 30 K and at low excitation powers of both wide and narrow QWs to preserve SC. The emitted light was collected in a cone of $\pm 0.15^{\circ}$. Under these conditions the PL from the lower and middle polariton 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.

The detuning dependence was performed under a fixed moderate n_e in the wide QW and at a temperature of 30 K in order to avoid any effects from charged exciton states. For each illumination spot, the measured photoluminescence enhancement $\Delta PL = [PL(I_X + I_n) - PL(I_n)]$ was normalized to the absorbed power of the wide QW, and $\eta = 100\%*\Delta PL/[PL(I_X) + PL(I_n)]$ was calculated. 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. The detuning dependence of the PL enhancement follows the same trend independently of the temperature, 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.



Fig. 3 Photoluminescence spectra for different free electron densities n_e (n_e increases upwards).

4 Electron density dependence

In order to reveal the extent of this enhancement, ΔPL was measured 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 normally-emitted PL spectra were recorded in figure 3 as n_e increases (upwards). Here the weak Ti:Sapphire pump, $I_X = 20$ mWcm⁻², only excites exciton densities of $n_X \approx 4*10^9$ cm⁻² (assuming $\tau_X \approx 500$ ps).



Fig. 4 (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 .

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. 4a), while the PL from the middle polariton (MP) mode conversely decreases. However, increasing n_e also systematically broadens the MP mode and red shifts its peak energy position, Fig.4b. 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 note that the red shift of the MP mode energy

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towards the bare X_{hh} energy marks a different critical diode power, I_n^{WC} , at which the system passes from the 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 the shape of the LP branch between the SC and WC regimes at these negative detunings Fig. 1a. 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 4meV), the transition to the WC regime occurs for moderate exciton or electron densities and therefore without substantially collision-broadening the exciton transition, so that excitons remain well defined states. Electron-exciton scattering thus exhibits similar properties in both the SC and WC regimes. 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, $n_e > 10^{11} \text{ cm}^{-2}$.



Fig. 5 PL enhancement vs. I_n for $\Delta = -1.5$ 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).

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. The PL enhancement with I_n shown in figure 5 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 behaviour 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.

A number of approximations are made in this model. The present phenomenological approach neglects the effects of bound charged excitons 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. In addition, the effects of both exciton and carrier localization have been ignored. This is a valid approximation for samples that are of high quality. In particular, the sample under investigation has polariton linewidths eight (8) times smaller than the Rabi splitting, which justifies the approximation.

5 Conclusions

We have presented evidence for the effectiveness of electrons within the active region of a stronglycoupled semiconductor microcavity, for enhancing relaxation of polaritons from the bottleneck region to the ground polariton state. This matches recent theoretical predictions. Understanding the ways that electron-exciton Coulomb scattering contributes differently to exciton broadening and photoluminescence enhancement paves the way to improved polariton emitters and lasers.

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