

Exciton warming in III–V semiconductors and microcavities

A. Amo^{a,*}, D. Ballarini^a, D. Sanvitto^a, E. Kozhemyakina^a, L. Viña^a,
M.S. Skolnick^b, J.S. Roberts^c

^a SEMICUAM Departamento de Física de Materiales, Universidad Autónoma de Madrid, E-28049 Madrid, Spain

^b Department of Physics, University of Sheffield, S3 7RH, Sheffield, United Kingdom

^c Department of Electronic and Electrical Engineering, University of Sheffield, S1 3JD, Sheffield, United Kingdom

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Abstract

We present a time resolved experiment in which we dynamically tailor the occupation and temperature of a photogenerated exciton distribution in QWs by excitation with two delayed picosecond pulses. The modification of the excitonic distribution results in ultrafast changes in the PL dynamics. Our experimental results are well accounted by a quasiequilibrium thermodynamical model, which includes the occupation and momentum distribution of the excitons. We use this model and the two-pulse experimental technique to study the polariton dynamics in InGaAs-based microcavities in the strong coupling regime. In particular, we demonstrate that resonantly injected upper polaritons mainly relax to the lower polariton branch via scattering to large momentum polariton states, producing the warming of the polariton reservoir.

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The prospect of the realization of a polariton laser [1] (*plaser*) based on semiconductor microcavities has risen much attention lately [2]. Optical emission in such a *plaser* would come from leakage out of the microcavity of a quantum condensate of polaritons in momentum (K) space at the energy minimum of the lower polariton branch. In the *plaser* the stimulation process takes place in the relaxation of reservoir lower polaritons with large K to $K = 0$ states at the bottom of the lower polariton branch. Once there, a macroscopic condensate of

* Corresponding author.

E-mail address: alberto.amo@uam.es (A. Amo).

polaritons can form in a well defined quantum state. The polaritons from this condensate that leak out of the cavity present very similar characteristics to those of a conventional photon laser, i.e. monochromaticity, well defined phase, directionality. However, the fact that the stimulation and emission processes are decoupled can lead to *plasing* devices without excitation threshold.

The realization of a practical *plaser* relies on the possibility of operation, under non-resonant excitation, in the regime of stimulated scattering [3] while preserving the strong exciton–photon coupling in the system. This situation has been obtained in microcavities based on II–VI materials and coherent emission from a polariton condensate has been demonstrated [2]. Although III–V GaAs-based structures would be of greater interest due to their high crystalline quality, the reduced exciton oscillator strength and the bottleneck effect [4] prevent the system from reaching the stimulated scattering regime while preserving the strong coupling under non-resonant excitation [5,6]. Several configurations have been attempted in order to reduce the bottleneck effect and enhance the polariton relaxation in the lower branch in these materials: (i) the increase of the lattice temperature to relax the energy–momentum conservation constrictions [4,7,8], and (ii) the introduction of free carriers in the system to enhance the polariton relaxation scattering [7,8]. However none of these approaches result in $K = 0$ occupations high enough to trigger the stimulated scattering processes. In this communication we investigate a new approach based on a non-resonant, two-pulse experiment. The first laser pulse would create a polariton population in the lower branch. Some polaritons would relax to $K = 0$ states and they would act as a seed for the stimulated relaxation of polaritons non-resonantly injected by a second pulse some hundreds of picoseconds after the arrival of the first pulse. We will show that the polaritons injected by the second pulse do not suffer stimulation processes to $K = 0$ states. Instead, the effect of the arrival of the second pulse is the warming of the polariton reservoir (i.e. polaritons with momentum larger than those at the bottleneck). The most striking result of this effect is the ultrafast drop of the lower polariton branch photoluminescence (PL) when the second pulse arrives. We will present a thermodynamical model and experiments in quantum wells that fully explain those processes. In the last paragraphs of this communication we will show some experiments in which we make use of this two-pulse configuration and the above mentioned model to study the relaxation from the upper polariton branch to the reservoir states.

The samples used for these studies were $3\lambda/2$ microcavities grown by metal–organic phase epitaxy, with two sets of three $\text{In}_{0.06}\text{Ga}_{0.94}\text{As}$ QWs at the antinodes of the electromagnetic field. The QWs are embedded in a GaAs spacer which is wedged, allowing the selection of the exciton–cavity energy detuning (δ) by exciting in different points in the sample. Above and below the cavity spacer, $\text{Al}_{0.1}\text{Ga}_{0.9}\text{As}/\text{GaAs}$ distributed Bragg reflectors were grown. A Rabi splitting of 6.6 meV between the upper and lower polariton branches was measured in a low power cw characterization at 5 K. We have also used GaAs/AlAs multiple quantum wells with a width of 70 Å.

Both the microcavities and the GaAs/AlAs multiple QWs were kept at 5 K and photoexcited with pulses 2 ps long. The pulses, both with the same energy, reached the sample in consecutive pairs whose power, polarization and delay was controlled. We focus on the emission from $K = 0$ lower branch polaritons, which was selected by means of a pinhole. The luminescence was energy and time resolved using a spectrograph in conjunction with a streak camera.

Fig. 1(a) shows the PL emission from the microcavity at $\delta = 0$ under non-resonant photoexcitation (180 meV above the exciton resonance). The dashed lines depict the PL after individual excitation by each of the pulses, which are delayed by 410 ps. When the two pulses reach the sample consecutively (solid red line) an abrupt decrease of the PL from the $K = 0$ polaritons is observed. The ultrafast decrease takes place at the time of the arrival of the second

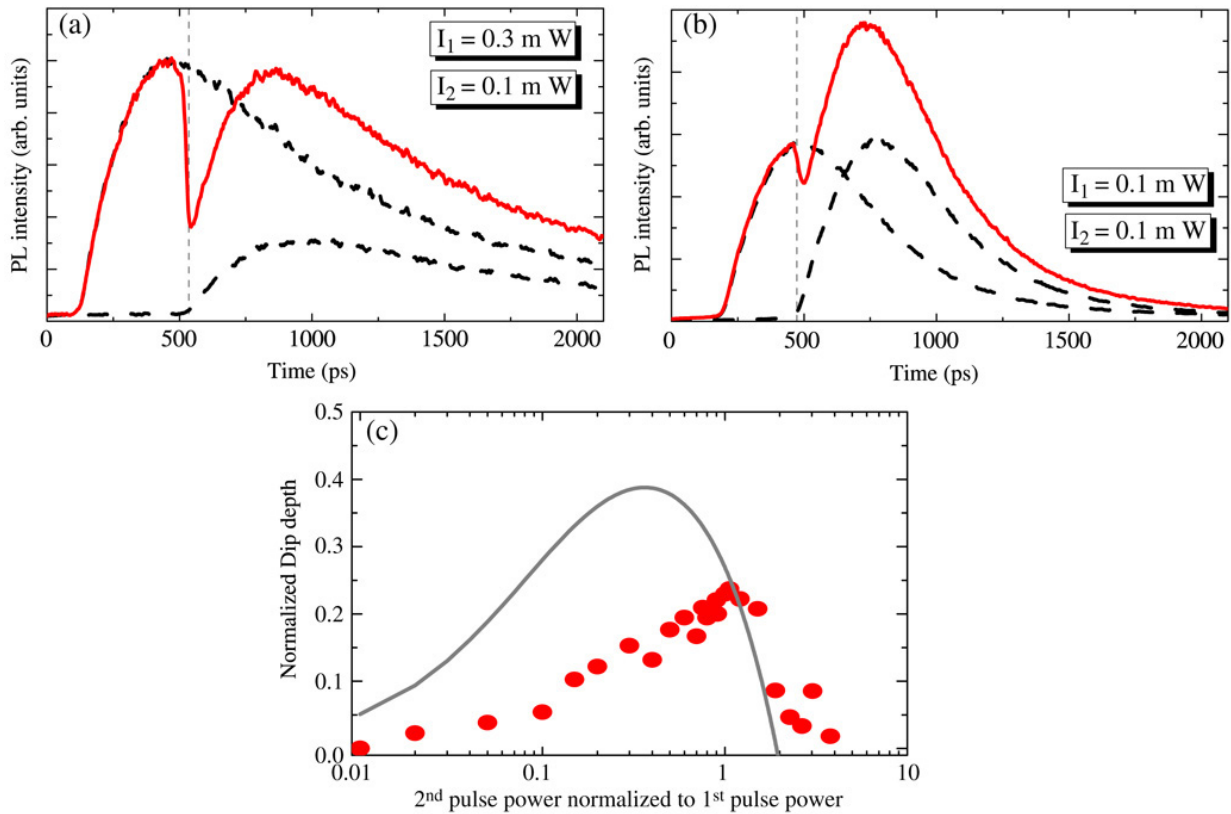


Fig. 1. (a) Microcavity time resolved PL of the $K = 0$ lower branch polaritons under non-resonant excitation at $\delta = 0$ for one-pulse experiments (dashed lines) and the two-pulse experiment (solid red line). (b) Same as (a) for the excitonic emission from GaAs/AlAs QWs. (c) Dip depth normalized to the PL intensity in the QWs right before the arrival of the second pulse as a function of the relative power of the second pulse (power of the first pulse: 0.1 mW); the solid line shows the results of the model as described in the text. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

pulse, and it is present for all the delays and powers studied. The observation of a dip instead of an increase in the PL, indicates that the $K = 0$ population created by the first pulse is not high enough to initiate the stimulated relaxation of the polaritons injected by the second pulse. This is a direct consequence of the short lifetime of $K = 0$ polaritons in the microcavity under study, which is of the order of 4 ps and disables any possibility for the build up of a high enough polariton population in these states to trigger stimulated processes.

In order to clarify the origin of the dip in the PL we have performed similar experiments on the GaAs/AlAs multiple QW samples. Fig. 1(b) depicts the time evolution of the free exciton PL for non-resonant excitation of the QW for each of the pulses individually (dashed lines) and for the two-pulse configuration (red solid line). A dip in the PL analogous to that observed in the microcavity experiments is also obtained, indicating that the origin of the dip can be entirely attributed to excitonic processes. The dependence of the dip depth with power of the second pulse has been measured and it is represented in Fig. 1(c).

We have developed a quasiequilibrium thermodynamical model to explain the origin of the dip in the PL and to quantify its depth in the QW experiments. When the first non-resonant pulse impinges on the QWs sample, electrons and holes are created with an excess energy in the conduction and valence bands, respectively. In a subpicosecond timescale, due to the high efficiency of carrier–carrier scattering processes, both populations thermalize and acquire a well defined temperature, which is above the lattice temperature [9]. As the photoinjected densities

considered here are low compared to the degenerate situation, both electron and hole populations can be well described by Maxwell–Boltzmann distributions. As time evolves the interaction of carriers with phonons cools down the carrier distributions [10], and radiative recombination reduces the total density of carriers. When the second pulse reaches the sample, hot electron–hole pairs are photocreated. Again, in a subpicosecond timescale the newly injected carriers and the preexisting populations thermalize and achieve a common well defined temperature, which is higher than the carrier temperature right before the arrival of the second pulse. In this way, the temperature of the carriers in the QWs experiences an ultrafast change when the second pulse reaches the sample. In particular, the arrival of the second pulse, due to the change in the carrier distributions, results in an abrupt change of the carrier occupations at the bottom of the bands (carriers with $k = 0$). The same arguments that we have employed so far for electrons and holes can be applied to the QW excitons, which are in thermodynamic quasiequilibrium with the electron–hole populations [11]. Therefore, the arrival of the second pulse results in an abrupt change in the number of excitons with center of mass momentum $K \approx 0$. These are the optically active excitons whose PL emission is depicted in Fig. 1(b), and the dip that they show in the PL is a direct consequence of the redistribution of carriers in the bands when the second pulse impinges on the sample.

Fig. 1(c) show in solid lines the results of the computation of the model considering the recombination rate of excitons in the QWs, the laser excess energy and the carrier cooling time in a similar sample under analogous conditions [11].

In the microcavity, at $\delta = 0$, analogous effects take place for polaritons in the reservoir. These polaritons present a high excitonic content and their physics is very similar to that of QW excitons. The dip in the $K = 0$ polariton PL under non-resonant excitation reflects the changes that take place in the polariton population above the bottleneck [12] when the second pulse reaches the sample [see Fig. 1(a)].

We have taken advantage of our description of the physics behind the dip in the PL to study the relaxation of polaritons from the upper branch to the lower branch. We have performed an experiment in which a first linearly polarized pulse resonantly injects polaritons at the bottom of the upper branch (see Fig. 2(a)). These polaritons relax towards the reservoir states of the lower polariton branch where they conform a thermalized distribution [13]. Some of these polaritons relax towards $K = 0$ states where they escape from the cavity (see Fig. 2(b), dashed lines). When a second delayed pulse reaches the sample at the same energy (see Fig. 2(a)), the photocreated upper polaritons relax again to the reservoir states with an excess energy given by the difference between the upper polariton energy and the reservoir energy (bare excitonic energy), as indicated by Δ in Fig. 2(a). Fig. 2(b) depicts in solid red line the PL emission in the two-pulse experiment when excitation is resonant with the upper polariton branch. Again a dip in the PL can be observed indicating that the upper polaritons generated by the second pulse very efficiently relax towards the reservoir states changing significantly their distribution. Let us mention that additional experiments (not shown here) show that the depth of the dip increases when the detuning is increased. This is supported by our model as the final temperature of the reservoir polaritons after the arrival of the polaritons photocreated by the second pulse increases with Δ , and so does the dip depth.

In summary, we have shown that the distribution of photogenerated carriers in a QW can be precisely modified and controlled by ultrafast light pulses with the proper delay and power. The change in the carrier distributions is evidenced as an abrupt decrease in the PL when a second delayed pulse reaches the sample. These effects are also observed in the PL emission of lower branch polaritons in semiconductor microcavities under non-resonant excitation, due to

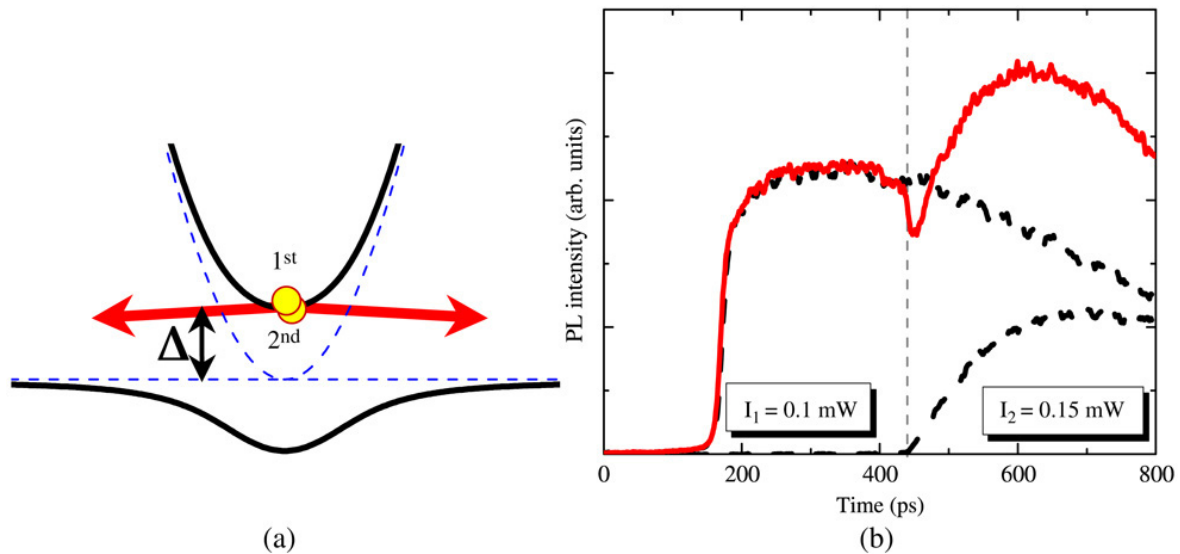


Fig. 2. (a) Solid lines: polariton dispersion in the microcavity for $\delta = 0$; dashed lines: bare exciton and cavity modes. (b) Microcavity time resolved σ^- PL of the $K = 0$ lower branch polaritons under resonant excitation in the upper polariton branch for one-pulse (dashed lines) and two-pulse experiments (solid red line). The first excitation pulse is linearly polarized, the second is σ^+ polarized.

the changes in the distribution of reservoir polaritons induced by the absorption of the delayed pulse. This effect has enabled us to study and demonstrate the efficient relaxation from resonantly photogenerated upper polaritons to reservoir lower polariton states.

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