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We have investigated the influence of polariton–phonon interaction on the relaxation of microcavity polaritons towards the bottom of the lower polariton branch (LPB). We have varied either the energy difference between the two polariton branches at $K \sim 0$ (negative detuning) or the Rabi splitting (zero detuning), adjusting them to coincide with the energy of a LO-phonon. We have observed a sub-linear/super-linear increase of the LPB emission with pump power for negative/zero detuning, respectively. These different power dependencies can be explained by the combined effect of the different polariton–phonon scattering efficiency, which is minimum/maximum for large negative/zero detunings, a competition between several relaxation mechanisms and the different coupling to the excitonic reservoir of large-K LPB states.

1 Introduction The possibility of achieving a Bose condensation in a semiconductor heterostructure has elicited great interest in the last decade, especially in the cases of quantum wells (QWs) [1] and microcavities (MCs) [2–4]. In MCs, evidences of a condensed state have been reported recently [4]. In these structures, it is possible to observe a condensation at moderately low temperatures ($\leq$20 K), due to the small mass of the polaritons ($\sim 10^{-5} m_0$) and the reduced density of states. This fact opens up the possibility of exploiting all the quantum effects related with a condensed state and integrating them into a semiconductor chip, eventually speeding up the development of quantum computing.

So far, several methods to obtain a macroscopic occupation of the ground state in semiconductor MCs have been considered. All of them are based in polariton–polariton scattering, which becomes stimulated by the ground-state population beyond a certain excitation power threshold, while maintaining the general polariton properties (i.e. without significant screening or bleaching contributions due to many-particle effects) [5–7]. This final-state stimulated scattering process results in a non-linear emission from the polariton ground state and an eventual condensation of polaritons. In this work we study the effect of resonant polariton–LO-phonon scattering on the relaxation of polaritons towards the ground state as an alternative to polariton–polariton stimulated scattering. To do so, we have adjusted the energy difference between the two polariton branches to coincide with the energy of a LO-phonon in CdTe. We have also exploited the possibility to change the predominant excitonic/photonic character of the polariton ground state offered by the MCs. We have considered first a mainly photonic ground state, setting the cavity mode below the exciton (negative detuning) while keeping the resonance with the LO-phonon. Then we studied the case of half exciton–half photon ground state, using another MC sample in which the characteristic Rabi splitting is comparable to the LO-phonon energy (zero detuning). In both cases, negative and zero detuning, we have compared the relaxation dynamics of polaritons after the well known case of non-resonant excitation (above the mirrors' stop-band) [8–10] and after resonantly creating polaritons on the upper polariton branch (UPB). We have found remarkable differences in both the relaxation and the emission dynamics for the two excitation conditions, even though in both cases one of the main channels for polariton

relaxation involves similar large-K lower polariton branch (LPB) states, in the vicinity of the relaxation bottleneck and the exciton reservoir.

2 Samples and experiments We have used two different MC samples. The first one, with a Rabi splitting at low temperature of 10 meV, is a $Cd_{0.4}Mg_{0.6}Te$ λ-MC with embedded 9 nm wide CdTe QWs. We have used this sample to study the negative detuning case, setting the cavity mode below the exciton in such a way that the energy difference between the UPB and the LPB coincides with that of a CdTe LO-phonon (21.3 meV). The second sample, with a Rabi splitting of 26 meV, is a $Cd_{0.4}Mg_{0.6}Te$ 2λ-MC containing 16 CdTe QWs of 50 Å width. We have used this sample to study the zero detuning case.

The measurements are performed at 5 K and the excitation beam arrives to the sample almost perpendicular to its surface, its energy fixed either above the mirrors' stop-band (non-resonant) or at the UPB (resonant). We have time-resolved the photoluminescence (PL) originating from LPB $K \sim 0$ states by means of a spectrograph coupled to a streak camera, with an overall timer resolution of ~15 ps.

3 Results and discussion

3.1 Negative detuning In this section we will discuss the results obtained for doubly-resonant Raman conditions after adjusting the exciton-cavity detuning to have a LO-phonon energy difference between UPB and LPB, selecting a polariton ground state with a predominant photonic character. Figure 1 displays the time evolution of the LPB $K \sim 0$ emission under non-resonant (a) and UPB resonant (b) excitation, for different pump powers. We observe the well know acceleration of the emission dynamics with power after non-resonant excitation (Fig. 1a). For these excitation conditions, the polariton relaxation process can be schematically described as follows [8]: the excitation pulse creates electron–hole pairs that bind to form hot excitons, which occupy the large-K states, on the LPB. These excitons thermalize via exciton–exciton scattering and approach the relaxation bottleneck. From there they reach $K \sim 0$ states via polariton–polariton scattering and/or acoustic phonon emission. Once the pump power is risen above a certain threshold, polariton–polariton scattering becomes stimulated by the population occupying the $K \sim 0$ LPB (final) states. This stimulation is confirmed by the super-linear dependence observed in the integrated emission (inset of Fig. 1a).

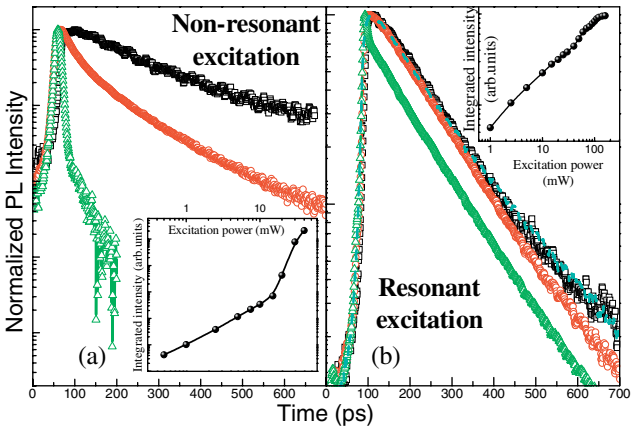

Figure 1 (online colour at: www.pss-b.com) (a) Semi-log plot of the normalized time evolution traces of the PL originating from $K \sim 0$ LPB states for a negative detuning resonant with a LO-phonon after non-resonant excitation, for excitation powers of 0.5/10/45 mW (open squares/circles/triangles). Inset: power dependence of the LPB integrated emission (log–log scale). (b) same as (a) but obtained after UPB resonant excitation. The pump powers are 1/25/100 mW (open squares/circles/triangles). The dashed line corresponds to a time evolution trace obtained after non-resonant excitation with 5 mW pump power.

Let us now turn to the polariton dynamics after resonant excitation on the UPB (Fig. 1b). The resonantly created polaritons have two main channels to relax to $K \sim 0$ LPB states [11]. The first channel (mechanism A) involves the direct relaxation to the LPB emitting a LO-phonon of negligible wave-vector. The second channel (mechanism B) requires two UPB polaritons scattering to opposite sides of the exciton reservoir region, on the LPB. From there, the relaxation would go on in a similar way as for non-resonant excitation, via polariton–polariton scattering and/or acoustic phonon emission [12].

We have extracted the rise and decay times of the emission using a simplified rate equation model. We have found a very short rise time (~15 ps) that does not change with excitation power, which is an evidence of a fast but inefficient relaxation to the LPB through mechanism A. In fact, the efficiency of polariton-phonon scattering is very small for negative detuning [13]. Only the decay time is slightly reduced increasing the pump power (125 to 110 ps). Such a long decay time is generally attributed to the relaxation of polaritons along the LPB via polariton–polariton scattering and/or acoustic phonon emission. A closer look to these time evolution traces reveals the similarity with those obtained in the linear regime after non-resonant excitation (dashed line in Fig. 1b). Furthermore, there is no trace of stimulated emission, as confirmed by the power dependence of the integrated PL (inset of Fig. 1b). This inhibition of the final-state polariton–polariton stimulated scattering could arise from two very different causes. The first one is the large reflectivity of the UPB at large negative detunings, such as this discussed here. The polariton population created by the effective excitation power that gets into the MC is not large enough to start the stimulated scattering processes. However it will not explain why the integrated emission dependence on pump power remains linear over more than two orders of magnitude. The second possible cause is that, as a result of the predominance of relaxation mechanism B, the scattering to large-K LPB states occur to a particular region, different from that reached after non-resonant excitation, from which polariton–polariton stimulated scattering is strongly inhibited, leading to a weaker emission from

$K \sim 0$ LPB states. Recently, remarkable differences between the nature of the observed $K \sim 0$ LPB stimulated emission after non-resonant/UPB-resonant excitation have been reported for zero detuning in III–V MCs [14], revealing that excitons and photons are strongly/weakly coupled, respectively, even though in the predominant relaxation channel similar intermediate large-K LPB states are involved in both cases. This is an evidence of the different exciton–polariton interaction of the intermediate LPB states. A similar effect could be happening in our MC: if the states reached via mechanism B and those after non-resonant excitation are distinct, they could interact differently with the exciton reservoir and display different relaxation dynamics.

3.2 Zero detuning In this section we will discuss the results obtained for doubly-resonant Raman conditions using a MC sample characterized by a Rabi splitting comparable to the energy of a LO-phonon. In this way we are selecting a polariton ground state with a half-exciton/half-photon character. For this detuning conditions the polariton–phonon scattering efficiency is maximum [13] and one can expect mechanism A to play a significant role in the polariton relaxation towards $K \sim 0$ LPB states. Figure 2 shows the normalized time evolution of the LPB $K \sim 0$ emission under non-resonant (a) and UPB resonant (b) excitation, for different pump powers. The emission dynamics are very similar for both excitation conditions: we observe an analogous acceleration of the recombination dynamics with power and a clear non-linear emission originating from $K \sim 0$ LPB states, evidenced by the pump power dependence of the integrated emission intensity (insets of Fig. 2). However, there are several subtle differences between the results obtained in each particular case, the first of which is the slightly slower emission dynamics found for the resonant excitation case. Let us compare, for example, the decay time extracted from the time evolution traces. In the case of non-resonant excitation we obtain a decay time of ~160 ps below the stimulation threshold that progressively shortens increasing the excitation power, being limited by the time resolution of the setup at large powers (~15 ps). For UPB-resonant excitation, the characteristic decay time below the stimulated emission threshold is ~200 ps, decreasing to ~25 ps above the threshold. A second difference between the dynamics found for the two excitation conditions is in the value of the pump power threshold required to observe the non-linear emission, which is one order of magnitude larger for the UPB-resonant excitation case. This slightly slower emission dynamics and the larger stimulation threshold found for the UPB-resonant case could originate from several causes: (i) a competition between mechanisms A and B, as they both feed from the same UPB population; (ii) the larger reflectivity of the cavity together with the lower absorption coefficient after resonant excitation at the UPB; (iii) as described in the previous section regarding the relaxation mechanism B, it could happen that the intermediate states reached after UPB pair scattering to large-K LPB states are different from those reached after non-resonant excitation. These different intermediate states will have a different coupling to the excitonic reservoir, leading to a delay of the relaxation along the LPB and a different threshold for the activation of polariton–polariton final state stimulated scattering.

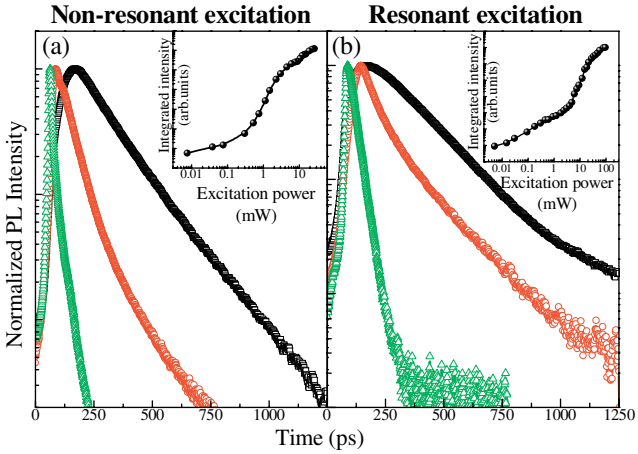

Figure 2 (online colour at: www.pss-b.com) (a) Semi-log plot of the normalized time evolution traces of the PL originating from $K \sim 0$ LPB states for zero detuning, close to resonance with a LO-phonon, after non-resonant excitation, for excitation powers of 0.7/1.5/30 mW (open squares/circles/triangles). Inset: power dependence of the LPB integrated emission (log–log scale). (b) same as (a) but obtained after UPB resonant excitation. The pump powers are 2/20/100 mW (open squares/circles/triangles).

4 Conclusion We have studied polariton-phonon scattering by means of time-resolved PL spectroscopy under doubly-resonant Raman scattering conditions as an alternative to obtain a non-linear emission from the LPB and eventually a macroscopic occupation of the polariton ground state. We have considered polaritons with different photonic/excitonic content, varying the exciton-cavity detuning while keeping the resonance with the LO-phonon. We have also taken into account different excitation conditions, creating polaritons both resonantly (at the UPB) and non-resonantly, and compared the results obtained for both excitations.

For the case of negative detuning, we have found that, after resonant excitation at the UPB, the direct relaxation to the LPB is strongly inhibited because of the small efficiency of polariton–phonon scattering for negative detunings, so that the predominant relaxation mechanism is the pair scattering of UPB polaritons to intermediate, large-K, LPB states, followed by a further relaxation along the LPB via acoustic phonon emission. In this case, polariton–polariton stimulated scattering appears to be inhibited although the intermediate states are similar to those reached after non-resonant excitation, suggesting that the interaction of these states with the excitonic reservoir is different.

For the case of zero detuning, we have found the relaxation and the emission dynamics to be very similar to those obtained after non-resonant excitation, with an acceleration of the relaxation increasing the pump power and a clear threshold for the observation of a non-linear emission, indicating that, in this case, polariton–polariton final-state stimulated scattering is not inhibited after UPB-resonant excitation. For this detuning, the polariton–phonon scattering efficiency is maximum. A competition between these two scattering mechanisms could explain the slight differences observed in the dynamics.

Further experiments are underway to identify the intermediate large-K LPB states from which UPB resonantly created polaritons relax to $K \sim 0$ LPB, so they can be compared to those reached after non-resonant excitation. Polariton–phonon scattering will also be studied for a mainly excitonic LPB, setting the LO-phonon resonance for positive detuning.

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References

[1] L. V. Butov, A. C. Gossard, and D. S. Chemla, Nature **418**, 751 (2002).
[2] M. Richard, J. Kasprzak, R. Romestain, R. André, and Le Si Dang, Phys. Rev. Lett. **94**, 187401 (2005).
[3] H. Deng, D. Press, S. Götzinger, G. S. Solomon, R. Hey, K. H. Ploog, and Y. Yamamoto, Phys. Rev. Lett. **97**, 146402 (2006).
[4] J. Kasprzak, M. Richard, S. Kundermann, A. Baas, P. Jeambrun, J. M. J. Keeling, F. M. Marchetti, M. H. Szymańska, R. André, J. L. Staehli, V. Savona, P. B. Littlewood, B. Deveaud, and Le Si Dang, Nature **443**, 409 (2006).
[5] P. G. Savvidis, J. J. Baumberg, R. M. Stevenson, M. S. Skolnick, D. M. Whittaker, and J. S. Roberts, Phys. Rev. Lett. **84**, 1547 (2000).
[6] R. M. Stevenson, V. N. Astratov, M. S. Skolnick, D. M. Whittaker, M. Emam-Ismail, A. I. Tartakovskii, P. G. Savvidis, J. J. Baumberg, and J. S. Roberts, Phys. Rev. Lett. **85**, 3680 (2000).
[7] M. Saba, C. Ciuti, J. Bloch, V. Thierry-Mieg, R. Andre, Le Si Dang, S. Kundermann, A. Mura, G. Bongiovanni, J. L. Staehli, and B. Deveaud, Nature **414**, 731 (2001).
[8] A. Alexandrou, G. Bianchi, E. Péronne, B. Hallé, F. Boeuf, R. André, R. Romestain, and Le Si Dang, Phys. Rev. B **64**, 233318 (2001).
[9] A. I. Tartakovskii, M. Emam-Ismail, R. M. Stevenson, M. S. Skolnick, V. N. Astratov, D. M. Whittaker, J. J. Baumberg, and J. S. Roberts, Phys. Rev. B **62**, R2283 (2000).
[10] M. D. Martín, G. Aichmayr, L. Viña, and R. André, Phys. Rev. Lett. **89**, 077402 (2002).
[11] J. Bloch and J. Y. Marzin, Phys. Rev. B **56**, 2103 (1997).
[12] The long-lived effects on the relaxation of polaritons due to the interaction of LPB states with localised excitons have not been taken into account, since this interaction is strongly inhibited in the cases of negative/zero detuning considered here. See for example: D. M. Whittaker, Phys. Rev. Lett. **80**, 4791 (1998); J. J. Baumberg, A. Armitage, M. S. Skolnick, and J. S. Roberts, Phys. Rev. Lett. **81**, 661 (1998).
[13] A. Fainstein, B. Jusserand, and R. André, Phys. Rev. B **57**, R9439 (1998).
[14] D. N. Krizhanovskii, A. P. D. Love, D. Sanvitto, D. M. Whittaker, M. S. Skolnick, and J. S. Roberts, Phys. Rev. B **75**, 233307 (2007).

