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Dynamics of polaritons resonantly created at the upper polariton branch

M.D. Martín[a,*], D. Ballarini[a], A. Amo[a], L. Viña[a], R. André[b]

[a] *SEMICUAM, Dept. Física de Materiales, Universidad Autónoma de Madrid, E-28049 Madrid, Spain*
[b] *Lab. Spectrométrie Physique, Université Joseph Fourier, F-38402 Grenoble, France*

Abstract

We have studied the polariton relaxation dynamics in a CdTe microcavity at low temperatures after resonant excitation into the upper polariton branch (UPB). Initially, we have set a negative exciton–cavity detuning, such that the energy difference between the two polariton branches coincides with that of an LO phonon. Our experimental results reveal a sublinear dependence of the integrated emission from the lower polariton branch (LPB) with excitation power. This evidences not only an inefficient LO phonon mediated relaxation from the UPB to the LPB but also a substantial inhibition of polariton relaxation along the LPB. After that, we have progressively reduced the negative detuning, approaching the exciton–cavity resonance. Under these conditions it is possible to observe a nonlinear emission arising from $K \sim 0$ LPB-states similar to that observed after nonresonant excitation. Marked oscillations are present in the time evolution traces, with a period that does not depend on excitation power or detuning.

Keywords: Microcavities; II–VI Semiconductors; Polaritons; Relaxation dynamics

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1. Introduction

In the last years, the possibility of achieving a polariton condensation in the bottom of the dispersion relation has elicited great interest [1]. In this paper we study the relaxation dynamics of resonantly created polaritons at the bottom of the upper polariton branch (UPB) to the lower polariton branch (LPB) to find out whether these excitation conditions can lead

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* Corresponding author. Tel.: +34 914 975 025; fax: +34 914 978 579.
E-mail address: dolores.martin@uam.es (M.D. Martín).

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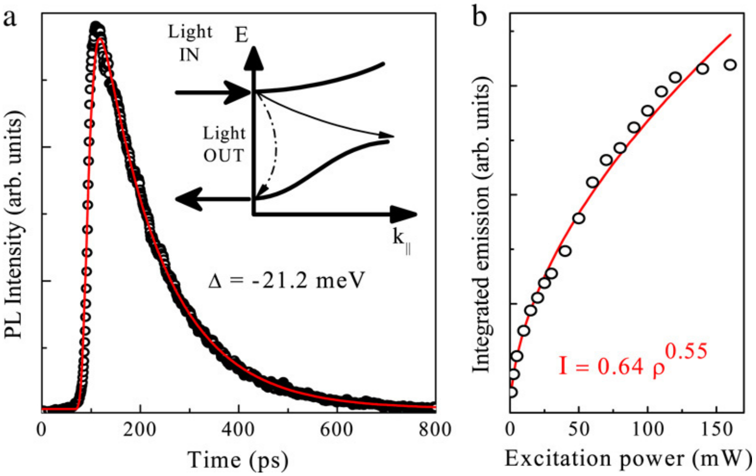

Fig. 1. (a) Time evolution trace of the emission from $K \sim 0$ LPB states for an excitation power $\rho = 1$ mW and $\Delta = -21.2$ meV. The line is a fit according to Eq. (1). Inset: schematic representation of the possible polariton–polariton scattering channels from the UPB. The energy- and K-conserving complementary channels are not depicted. (b) Integrated PL from $K \sim 0$ LPB states as a function of the excitation power. The solid line represents a fit to the power law shown in the graph.

to a macroscopic occupation of the LPB at $K \sim 0$. Previous reports in III–V microcavities indicate that direct scattering from $K \sim 0$ states in the UPB to $K \sim 0$ states in the LPB is inhibited [2,3] and show that UPB polaritons are more efficiently scattered to large-K LPB states. From these final (large-K) LPB-states, one could expect that the polariton relaxation would occur in a similar way as for nonresonant excitation, with a large polariton population accumulation at the bottleneck, followed by polariton–polariton stimulated scattering towards $K \sim 0$ for large enough excitation powers [4–6]. This stimulated scattering process results in a nonlinear emission arising from $K \sim 0$ LPB-states. However, our UPB-resonant experiments on the vicinity of the LO-phonon resonance show no evidence of such a non-linear emission, observing a remarkably sublinear emission. Only for smaller detunings a nonlinear emission similar to that obtained under nonresonant excitation is found.

2. Sample and experiments

The sample under study is a $Cd_{0.4}Mg_{0.6}Te$ λ-microcavity with embedded 9 nm wide CdTe quantum wells (characteristic Rabi splitting $\Omega = 10$ meV). The measurements are performed at 5 K and the excitation laser pulse is tuned to the UPB, arriving to the sample almost perpendicular to its surface. We have investigated several negative exciton–cavity detunings. Initially we have set an energy difference between UPB and LPB (Δ) close to the LO phonon energy of CdTe (21.2 meV). The polaritons resonantly created at $K \sim 0$ in the UPB can follow two different paths to recombination: they can be coherently reemitted at the UPB energy or they can relax to the LPB. To do the latter, they can either directly decay by emitting a LO phonon of negligible wave-vector or a pair of polaritons can scatter to opposite sides of the LPB dispersion relation (inset of Fig. 1(a)). We have time-resolved the photoluminescence (PL) originating from LPB-$K \sim 0$ states by means of a spectrograph coupled to a streak camera. For polarization-resolved measurements we have used two $\lambda/4$ plates: the excitation is σ^+-polarized and the PL is analysed into its σ^+- and σ^--polarized components.

3. Results and discussion

3.1. LO-phonon resonance

In this section we will present and discuss the results obtained for $\Delta = -\hbar\omega_{\mathrm{LO}}$. We have analysed the polariton relaxation dynamics by means of a simple rate equation model, which provides the rise (t_r) and decay (t_d) times of the LPB emission through a fit of the time evolution traces according to the expression

$$I \propto \left(\mathrm{e}^{-(t-t_0)/t_r} - \mathrm{e}^{(t-t_0)/t_d}\right). \qquad (1)$$

Fig. 1(a) displays a typical time evolution trace of the emission from the LPB for an excitation power of 1 mW (symbols) and a fit performed according to Eq. (1). We observe a very fast rise of the PL ($\leq$15 ps), which is not modified by the increase of excitation power over two orders of magnitude. This short t_r is related to a fast but inefficient relaxation to the LPB [2,3,7]. The situation could be different for $\Omega = \hbar\omega_{\mathrm{LO}}$, where a large enhancement of the phonon–polariton interaction is observed [7]. Only t_d is slightly affected changing the excitation power, decreasing from 125 to 110 ps. This time is much longer than the photon lifetime inside the cavity ($\sim$10 ps) and is generally attributed to the relaxation of polaritons along the LPB via the emission of acoustic phonons and/or polariton–polariton scattering.

One of our most remarkable findings is a complete absence of nonlinear emission from $K \sim 0$ LPB-states, which has been reported in an identical system under nonresonant excitation [5,6,8]. In fact, we have found a sublinear dependence of the integrated emission on excitation (Fig. 1(b)). This sub-linearity evidences an inhibition of polariton–polariton final-state stimulated scattering for these settings. This behaviour could be related to two very different causes. The first one is the large reflectivity of the microcavity at the UPB energy for large negative detuning, such as this. The effective excitation power that gets inside the cavity is much smaller than in the nonresonant case and therefore it is not possible to achieve polariton populations large enough to drive the system into the nonlinear regime. Yet it would not explain why the dependence is so clearly sublinear. The second possibility is the fact that $K \sim 0$ UPB polariton scattering to large-K states in the LPB occurs to a particular region, further from the bottleneck than in the case of non-resonant excitation, from which polariton–polariton scattering is strongly inhibited, resulting in a weaker emission from $K \sim 0$ LPB-states.

3.2. Smaller detunings

Only after a large reduction of the exciton–cavity detuning does the integrated emission from $K \sim 0$ LPB-states display a nonlinear dependence on excitation power, as seen in the inset of Fig. 2(a) for $\Delta = -13.8$ meV. Instead of a sublinear dependence ($\sim\sqrt{\rho}$ for $\Delta = \hbar\omega_{\mathrm{LO}}$, Fig. 1(b)) we find a quadratic increase of the maximum emission intensity with excitation power. This quadratic dependence on pump power evidences the efficiency of polariton pair scattering in the relaxation process (mechanism depicted in the inset of Fig. 1(a) with a solid line). As a result of such scattering process the LPB bottleneck population will acquire a quadratic dependence on pump power that will be inherited by the population at $K \sim 0$ as long as the coherence is maintained (short times and large pump powers).

Let us now describe the changes induced in the relaxation dynamics changing the pump power. Fig. 2(a) displays the time evolution traces of the σ^--polarized $K \sim 0$ LPB-states emission for $1/100$ mW (dotted/solid line) and $\Delta = -13.8$ meV. For small powers (dotted line) the LPB

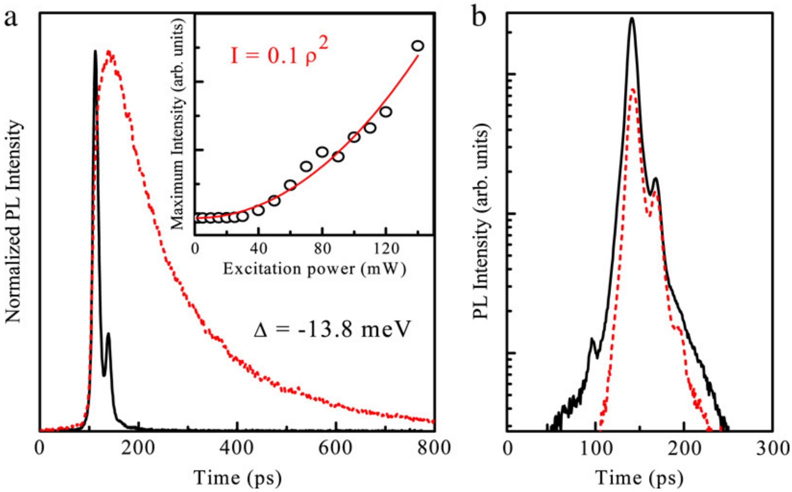

Fig. 2. (a) Normalized time evolution traces of the σ^--polarized emission from $K \sim 0$ LPB states for an excitation power, ρ, of 1/100 mW (dotted/solid line) and $\Delta = -13.8$ meV. Inset: maximum intensity of the σ^+-polarized emission from $K \sim 0$ LPB-states as a function of the excitation power. The solid line represents a quadratic fit. (b) Semilogarithmic plot of the time evolution traces of the σ^+/σ^--polarized PLs (solid/dotted line) for $\rho = 140$ mW and $\Delta = -13.8$ meV.

emission resembles that of the linear, nonresonantly excited PL with $t_d \sim 120$ ps. The increase of excitation power leads to a drastic acceleration of the dynamics (solid line) and the emission lasts barely 100 ps. Furthermore, increasing the pump power also results in the appearance of marked oscillations in the time evolution traces, with a characteristic beating time of 26 ps. This period corresponds to an energy difference of 160 μeV, which is much smaller than all the characteristic energies of the system. Additionally, we find that the oscillations of the +1 and −1 spin populations are in phase, since the σ^+- and the σ^--polarized emissions show maxima and minima for the same delay times (Fig. 2(b)). It should be made clear that these beatings occur spontaneously, as there is no external magnetic field applied. A similar behaviour has been previously reported [9]. It is particularly striking the excellent agreement between the beating period we observe here and that of Ref. [9], even though the excitation conditions are significantly different in both experiments. The beats are very robust: the beating period does not change with excitation power or exciton–cavity detuning. The oscillations can be attributed to the quantum beats between bright exciton–polaritons (with ±1 spin) and dark excitons (with ±2 spin) in the vicinity of the LPB bottleneck region [10]. The energy difference between bright and dark exciton states at the bottleneck will not change much with detuning or pump power. The only noticeable effect of the detuning is on the threshold excitation power from which the oscillations can be observed. The threshold power increases with $|\Delta|$. This could be a consequence of (i) a larger reflectivity of the cavity at the UPB energy increasing Δ (resulting in a smaller intra-cavity population) or (ii) the progressive inhibition of polariton–polariton scattering in the LPB for large detunings (the scattering from $K \sim 0$ UPB to large-K LPB occurs to states of too large wave-vectors, hindering the formation of a substantial population at the bottleneck from where to scatter down to $K \sim 0$).

4. Conclusions

We have studied the relaxation dynamics of polaritons resonantly created at the bottom of the UPB as a function of excitation power and exciton–cavity detuning. We have found that the efficiency of the relaxation from $K \sim 0$ UPB- to LPB-states is not strongly modified by setting $\Delta = -\hbar\omega_{\mathrm{LO}}$, at least for the negative detuning case considered here. The inhibition of polariton–polariton final-state stimulated scattering results in a sublinear dependence of the integrated emission on pump power. We have found a nonlinear emission from $K \sim 0$ LPB-states, similar to the one observed under nonresonant excitation, reducing Δ. It is possible to observe several oscillations of the emission intensity for large enough pump powers, with a beating period of 26 ps, which does not depend on detuning or power. The beats have been attributed to the coherent transfer of excitons between bright and dark states at the bottleneck. However, the resonant creation of polaritons in the UPB is not an advantageous alternative to nonresonant excitation, since the relaxation dynamics is still controlled by the relaxation bottleneck at large-K LPB-states.

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