Vol. **106** (2004) *ACTA PHYSICA POLONICA A* No. 3

Proceedings of the International Workshop "Optical Properties of 2D Systems", Warsaw 2004

Dynamics of Polariton Emission in the Linear Regime

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We studied the dependence of polariton emission dynamics on polariton wave-vector and exciton-photon energy detuning. To reproduce the experimental data, we applied a model that obtains the photonic and the excitonic emission rates of the polariton.

PACS numbers: 71.36.+c, 71.55.Gs, 78.47.+p

1. Introduction

The strong coupling between quantum well (QW) excitons and cavity photons leads to the appearance of new quasiparticles, named cavity polaritons [1]. The wave function of these particles is a linear combination of excitonic and photonic wave functions, and the excitonic and photonic content depends crucially on the detuning between bare exciton and cavity photon energies. Polaritons, being composed of bosons, manifest their bosonic nature in optical non-linearities based on quantum state occupancy. Due to their small mass, the photon-like polaritons have a very small density of states and their dispersion exhibits a large curvature. As a consequence, the probability of scattering with phonons is very low resulting in a "relaxation mobility edge" [2]. The experimental manifestation of this effect is a maximum in the emission intensity that is observed not at $k = 0$, but at larger values corresponding to the so-called bottleneck region. This bottleneck effect can be overridden by increasing the number of polaritons: above a certain excitation density threshold, the energy relaxation occurs via polariton–polariton scattering, stimulated by the final state occupancy [3]. Below this threshold, the

maximum emission comes from the states corresponding to the bottleneck region of the dispersion and its intensity depends linearly on the excitation density (linear regime). In this regime, the decay rates of non-resonantly excited polaritons are low, since the relaxation depends on the phonon scattering, which is not efficient. Above this threshold, the emission increases non-linearly with the pump power (non-linear regime) and the emission from $k = 0$ dominates. In this case, the decay rates are much larger than in the linear regime [4]. However, in the vicinity of the stimulated scattering threshold, both linear and non-linear regimes coexist, as seen in the decay profiles with two well defined characteristic times [4].

In this paper, we report on the polariton dynamics in the linear regime. Below the stimulated scattering threshold, the final state stimulation is weak which allows the investigation of dynamics of non-interacting polaritons. We describe our experimental results by a model where the excitonic and photonic dynamics are separated.

2. Experiment

The studied sample was a $Cd_{0.4}Mg_{0.6}Te$ λ microcavity with two CdTe QWs placed at the antinodes of the confined electromagnetic field. The top (bottom) cavity mirrors are distributed Bragg reflectors consisting of 17.5 (23) pairs of $Cd_{0.4}Mg_{0.6}Te/Cd_{0.75}Mn_{0.25}Te$ layers. The cavity is wedge-shaped, which allows us to vary the detuning between the bare exciton and cavity photon energies by changing the spot on the sample.

The emission was excited by an Al_2O_3:Ti laser giving 2 ps pulses pumped by an Ar^{2+} ion laser. The excitation power was kept below the stimulated scattering threshold to assure that the polariton dynamics are only weakly influenced by final state occupancy. To maintain the same excitation conditions for different detunings, the excitation energy was kept 62 meV above the photonic polariton, which corresponds to the first minimum of the cavity stop-band. The signal was detected and time-resolved by a synchroscan streak camera. The overall time resolution was 10 ps. To investigate the emission from various states along the po-

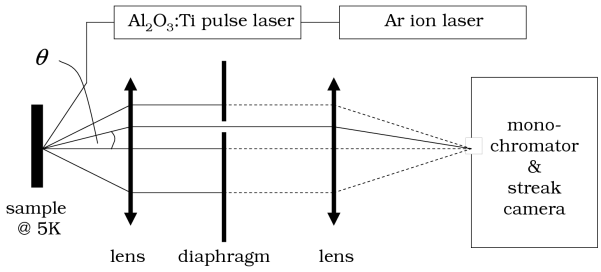

Fig. 1. Scheme of the experimental setup. θ is the angle between the measured beam and the normal to the cavity plane.

lariton dispersion curve, we measured the signal at various angles, θ, with respect to the normal to the sample surface. The experimental setup is schematically depicted in Fig. 1. We selected the emission angle by placing a diaphragm in a parallel signal beam. The relation between the emission angle and the parallel component of the polariton wave vector is given by: $k = \frac{E_{\rm ph}}{\hbar c} \sin\theta$, where $E_{\rm ph}$ is the energy of the bare cavity photon. The angular resolution was better than 1°, corresponding to a k-vector resolution of the order of 10^3 cm^{-1}.

The measurements were performed at the temperature of 5 K. Here, we present wave-vector dependent studies only for negative detunings, ranging from –20 to –5 meV, where the bottleneck effects are important.

3. Results

In the time-integrated spectra, three transitions are observed: the lower polariton (LP) and the upper polariton (UP) transitions resulting from the strong coupling between the excitons and photons, and a transition due to uncoupled bound excitons. From the energy positions of the LP and UP lines, we determined a Rabi splitting equal to 9.8 meV and obtained the detuning values for each spot of the sample.

In Fig. 2, the decay rates of the LP and UP at $k = 0$ are presented as a function of detuning. The LP decay is usually bi-exponential. We assume that the initial decay is due to a fast relaxation via the final state stimulation process [3, 4]. Therefore, in the following, we will discuss the decay rates observed at long delays. The UP decay rate increases by a factor of 6 as the detuning is increased from –20 to +30 meV. However, the LP rates show only a weak variation, they only decrease, by a factor of about 2, in the same detuning range.

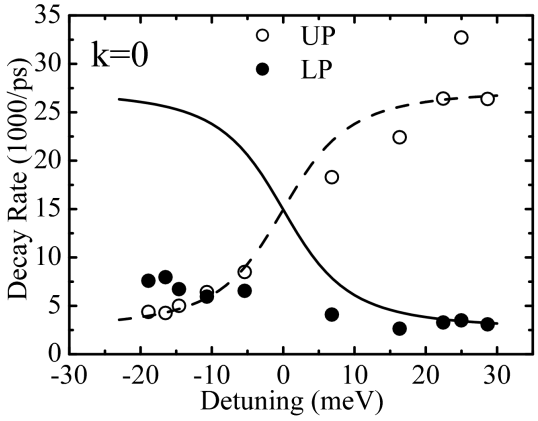

Fig. 2. Polariton decay rates measured for $k = 0$ at various spots on the sample corresponding to various exciton–photon detunings (see text). Open (full) points denote upper (lower) polariton branches. The dashed (solid) lines represent model calculation results for upper (lower) polaritons.

The dispersion curves of the LP and UP measured at the detuning of –14.6 meV are presented in Fig. 3. The UP transition is only seen within a small range around $k = 0$. The region where the maximum emission is observed (marked by an arrow) is located around 2.7×10^4 cm^{-1}, and its position does not change within the range of the investigated detunings.

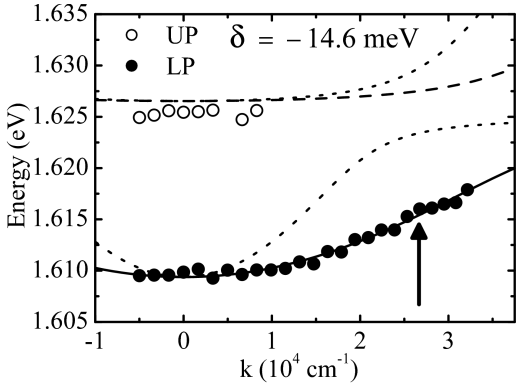

Fig. 3. Polariton energies measured as a function of the k vector. Open (full) points denote upper (lower) polariton branches. Dashed (solid) lines represent calculated upper (lower) polariton energies. The arrow marks the bottleneck region. Dotted lines are dispersion curves assuming the theoretical photon mass value (see text).

The solid and dashed lines in Fig. 3 are calculated as the eigenvalues of the Hamiltonian [5]:

$$H(k) = \begin{pmatrix} E_{\rm ph}(k) & \frac{1}{2}\hbar\Omega \\ \frac{1}{2}\hbar\Omega & E_X(k) \end{pmatrix}, \tag{1}$$

where k is the wave vector in the cavity plane, $E_{\rm ph}(k)$, $E_X(k)$ are the cavity photon and bare exciton dispersions, respectively, and Ω is the Rabi frequency. The exciton dispersion is assumed to be parabolic with an in-plane mass being a sum of the in-plane electron and hole masses. In turn, the photon dispersion is $E_{\rm ph}(k) = \hbar v\sqrt{k^2 + k_z^2}$, where v is the speed of light and the quantized light wave vector perpendicular to the cavity plane $k_z = m\pi/L_{\rm C}$, where $L_{\rm C}$ is the cavity length and m an integer. For small k (small angles) this dispersion can be approximated by

$$E_{\rm ph}(k) \approx \hbar k_z v + \frac{1}{2}\frac{vL_{\rm C}}{\pi\hbar}\hbar^2k^2 = E_{\rm ph}(k=0) + \frac{\hbar^2k^2}{2m_{\rm ph}}. \tag{2}$$

The photon "mass" $m_{\rm ph}$ is therefore given by the length of the cavity and the speed of light. However, if this mass is used for modelling the polariton energies, the calculated curvature is considerably larger than the one obtained from the measurement (see Fig. 3). Leaving $m_{\rm ph}$ as a fitting parameter we obtain a good fit of the experimental data, but the best fit values are by a factor of 3 to 6 larger,

depending on detuning, than the theoretical photon mass. Discrepancies between the experimental dispersion curves and predictions of Eq. (2) were also reported e.g. by Tartakovskii et al. In their case, the disagreement is ascribed to the fact that the emission intensity from $k = 0$ states is much larger than from states with small non-zero k. As a consequence, the strong emission masks the position of the small non-zero k states, resulting with flattening of the LP dispersion [6].

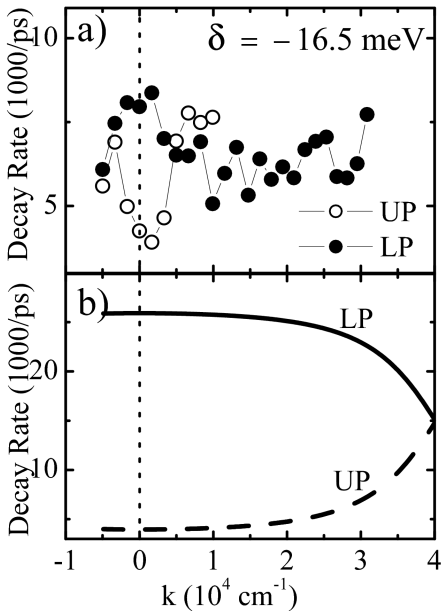

Fig. 4. (a) Upper and lower polariton decay rates measured as a function of the k vector. Open (full) points denote values for upper (lower) polariton branches. (b) Results of the model calculations. The dashed (solid) lines represent results for upper (lower) polaritons.

Figure 4a presents the decay rates measured as a function of the k vector at a detuning of –16.5 meV. A minimum and a maximum of the UP and LP decay rates are observed, respectively, at $k = 0$. At large k values, the UP transition disappears, while the LP decay rates, after an abrupt initial decrease remain constant within experimental accuracy.

4. Discussion

We model the polariton decay rates as

$$\frac{1}{\tau_{\mathrm{d}}} = \frac{a_X}{\tau_X} + \frac{a_{\mathrm{ph}}}{\tau_{\mathrm{ph}}}, \tag{3}$$

where $1/\tau_X$ and $1/\tau_{\mathrm{ph}}$ are excitonic and photonic decay rates, and a_X and a_{ph}

(the Hopfield coefficients) are excitonic and photonic parts of the polariton wave function, both dependent on k vector and detuning [7]. The above model proved to reproduce very well the decay rates of LP and UP under resonant excitation as a function of detuning, where emission was k-integrated around $k = 0$ [8]. The obtained photonic decay rate was in this case found to be exclusively related to the lifetime of the cavity photon.

The results of the model calculations as a function of detuning at $k = 0$ are presented as solid lines in Fig. 2. It is seen that the model reproduces very well the variation of the UP decay rates, while strongly overestimates those of the LP at negative detunings. We attribute this discrepancy to the existence of the relaxation bottleneck: the bottleneck slows down the relaxation to $k = 0$ and consequently makes the decay rates smaller. At positive detunings, where the influence of the bottleneck is negligible, the model agrees very well with the experimental results. Fitting the model to the experimental data we obtained characteristic times of the cavity photon and the bare exciton equal to 37 ps and 390 ps, respectively. These values are significantly larger than exciton and photon radiative times, but remain in good agreement with non-resonant excitation studies reported previously [9].

The situation becomes more complicated as k is increased. It is seen from Fig. 4a that the UP decay rates strongly increase with increasing k (up to 1×10^4 cm^{-1}). We attribute this behavior to the opening of another relaxation channel for UP: at $k = 0$ the relaxation from UP to LP is strongly inhibited as the momentum and energy conservation requires an intermediate state and eventually a two-phonon scattering process. Therefore, the UP decay rate not only reflects the radiative decay, but also relaxation from UP to LP states. For $k > 1 \times 10^4$ cm^{-1} this relaxation becomes so fast that the UP emission disappears. The calculations by Tassone et al. [10] and Savona et al. [11] based on a rate equations approach and diagonalization of full quantum mechanical Hamiltonian, respectively, predict a sharp minimum of radiative rates of UP at $k = 0$.

On the other hand, the LP decay rates decrease slightly as k is initially increased. The maximum at $k = 0$ probably reflects the existence of a weak final state stimulation process, which would dominate the emission dynamics as the excitation density is further increased. The models from Refs. [10] and [11] also predict this maximum, but the calculated decrease in the radiative rates is much more pronounced than the one found in our experiments.

Our results disagree with those of Müller et al., who observe a distinct increase in the LP decay rates with increasing k [2]. They explain their result by consequence of two relaxation paths: one — intrinsic — via phonon scattering along the polariton dispersion curve, and the other — extrinsic — via relaxation through bound exciton states. According to their interpretation, the small decay rates around $k = 0$ are due to relaxation via the latter path, as the former is practically forbidden by energy and momentum conservation. In our case the density of bound exciton states is considerably larger (as borne out by experiments not

discussed here), which in turn leads to faster relaxation via the extrinsic path. Indeed, the emission intensity from the bottleneck region is comparable to that from $k = 0$ (not shown).

Figure 4b presents the UP and LP decay rates, τ_{d} (see Eq. (3)), calculated as a function of k at the detuning of –16.5 meV with τ_X and τ_{ph} obtained from the detuning dependence fit. The k dependence of τ_{d} arises from the dependence of the Hopfield coefficients. It is clearly seen that in the whole range of k vectors the LP decay rates are strongly overestimated. One would expect that above the relaxation bottleneck the model will reproduce the experimental data, but this is not the case. We conclude that a proper description of the LP dynamics requires a more elaborate approach as for example that used in Refs. [10] and [11]. Furthermore, the scattering from the bottleneck reservoir to the states below and above in energy should also be taken into account. This ensemble of dynamical processes should lead to a substantial decrease in the decay rates with respect to the predictions of the simple model.

5. Conclusions

We obtained the decay rates for the upper and lower polariton branches as a function of exciton–photon detuning and polariton wave vector in semiconductor microcavities. A model based on separation of exciton and photon dynamics by means of the Hopfield coefficients describes well the upper polariton dynamics at $k = 0$. It is also inferred that at larger k the relaxation between polariton branches plays an important role in the polariton dynamics. We find that lower polariton dynamics are governed by the existence of a relaxation bottleneck. For a quantitative description of the decay rates, intra- and inter-branch relaxation processes have to be included in the description.

Acknowledgments

This work was partially supported by the Spanish MCYT (MAT2002-00139), the CAM (07N/0042/2002) and the "Marie-Curie" MRTN-CT-2003-503677 "Clermont2" ("Physics of Microcavities").

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