Polariton relaxation dynamics in semiconductor microcavities: Non-Markovian effects

F.J.Rodríguez*, L.Quiroga*, C.Tejedor†, M.D.Martín** and L. Viña**

*Departamento de Física, Universidad de los Andes, A.A. 4976, Bogotá D.C., Colombia
†Departamento de Física Teórica de la Materia Condensada, Universidad Autónoma de Madrid, Cantoblanco, E-28049, Madrid, Spain
**Departamento de Física de Materiales C-IV, Universidad Autónoma de Madrid, Cantoblanco, E-28049, Madrid, Spain

Abstract. We report results on the theoretical study of the ultrafast relaxation of polaritons in semiconductor microcavities. The photoluminescence decay dynamics is found to be controlled by the initial density of excitons. In the high density regime, the parametric conversion of excitons dominates the relaxation with memory (Non-Markov) effects producing a non-monotonous decay of the emission signal. This behavior agrees with recent experimental observations.

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Semiconductor microcavities have attracted a great deal of attention in recent years due to the possibility of tailoring the matter-radiation interaction in a solid-state environment [1, 2]. Recent results on time-resolved polariton photoluminescence (PL), following a non-resonant pump pulse exciting a quasi-two-dimensional II-VI semiconductor quantum well microcavity, show unusual features challenging conventional theoretical approaches [3, 4]. In particular, it has been reported that a very different emission dynamics is observed when an initial high exciton density is created by a high intensity laser pulse. Unexplained oscillatory/plateaux time behavior in PL decay is clearly present in the high excitation regime [4, 5]. Furthermore, spin effects could manifest themselves in experiments where co- and counter-polarized emissions are analyzed.

Most theoretical models describe the polariton dynamics within the Born-Markov approximation, which is indeed adequate for weak couplings and/or the long-time limit. In the present work we show that oscillations in the time-resolved emission signals that memory (non-Markovian) effects in the polariton evolution from an initially pumped bath of excitons relaxing towards $\overrightarrow{k} \sim 0$, the optically active polariton states, are present. The intensity dependent dynamics can be explained by resorting to an effective model where polaritons are coupled to an exciton bath. The coupling strength is determined by the excitation power. Possible spin effects are not included in the present study.

By the parametric down conversion process two excitons are simultaneously scattered: one of them goes to a near $\overrightarrow{k} = 0$ polariton state in the lower polariton (LP) branch while the other one (conserving energy and linear momentum) goes to a high $|\overrightarrow{k}|$ exciton state, the so-called idler particle. The Hamiltonian can be described as ($\hbar = 1$)[6, 7]

$$H = \sum_{\vec{q}} E(\vec{q}) a_{\vec{q}}^{\dagger} a_{\vec{q}} + \sum_{\vec{k}} \omega(\vec{k}) b_{\vec{k}}^{\dagger} b_{\vec{k}} + H_{XP} + H_{SI} \qquad (1)$$

where $E(\vec{q})$ describes the LP bare energy while $\omega(\vec{k})$ corresponds to the bare exciton energy (polaritons and excitons are described by boson-like operators a and b, respectively). The polariton-exciton interaction term is given by $H_{XP} = \sum_{\vec{k},\vec{k}',\vec{k}_1,\vec{k}_2} \tilde{V}(\vec{k},\vec{k}',\vec{k}_1,\vec{k}_2) a_{\vec{k}}^{\dagger} b_{\vec{k}'}^{\dagger} b_{\vec{k}_1} b_{\vec{k}_2} + H.C.$ where $\tilde{V}(\vec{k},\vec{k}',\vec{k}_1,\vec{k}_2)$ accounts for the polariton-exciton coupling and $H.C.$ means hermitic conjugate. The self-interaction term associated to the condensed $\overrightarrow{k} = 0$ polariton state is given by $H_{SI} = V_0 a_0^{\dagger} a_0^{\dagger} a_0 a_0$.

We consider that excitons optically pumped out of resonance rapidly relax into excited polaritons. At a high intensity pumping the initial density of excited polaritons can be described by classical fields, the so called parametric approximation. This approximation ignores quantum fluctuations in the pump polariton fields. Pump depletion is well accounted for with a pulse shaped classical field as determined by the shape/length of the excitation laser pulse as well as by proper exciton relaxation mechanisms in a quantum well. In H_{XP}, the $b_{\vec{k}_1} b_{\vec{k}_2}$ operator is replaced by the c-number $< b_{\vec{k}_1} b_{\vec{k}_2} > \sim \mathscr{I}_{\vec{k}_1}(t) \delta_{\vec{k}_1,\vec{k}_2}$ with $\mathscr{I}_{\vec{k}}(t)$ the effective pump intensity envelope. Thus, the polariton-exciton interaction term adopts the time-dependent effective form

$$H_{XP}(t) = \sum_{\vec{k},\vec{k}'} V(\vec{k},\vec{k}',t) a_{\vec{k}}^{\dagger} b_{\vec{k}'}^{\dagger} + \sum_{\vec{k},\vec{k}'} V^*(\vec{k},\vec{k}',t) a_{\vec{k}} b_{\vec{k}'} \qquad (2)$$

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where $V(\vec{k},\vec{k}',t)=\sum_{\vec{k}_1,\vec{k}_2}\tilde{V}(\vec{k},\vec{k}',\vec{k}_1,\vec{k}_2)\mathscr{I}_{\vec{k}_1}(t)\delta_{\vec{k}_1,\vec{k}_2}$. Note that in Eq.(2), the Hamiltonian describing the coupling of LP particles ($a_{\vec{k}}$ modes) to a high energy exciton bath ($b_{\vec{k}}$ modes), the coupling strength $V(\vec{k},\vec{k}',t)$ is now dependent on the excitation pulse intensity. This is why a Born-Markov based theoretical approach should fail: (i) even in the weak coupling regime a time-dependent coupling strength yields to a nonexponential decay and (ii) a strongly coupled system-reservoir is achieved beyond a certain excitation power. Furthermore, it is worth noting that the Hamiltonian in Eq.(1), with H_{XP} as given by Eq.(2), corresponds to a nondegenerate parametric amplifier for massive particles, where the parametric gain implies that the LP population, as well as exciton bath population, is amplified at the expenses of pump polaritons depletion. Since we are considering a pulsed situation, no indefinite gain will arise and the validity of the parametric approximation is still guaranteed.

The effective time-dependent polariton-bath coupling is taken as $V(\vec{k},\vec{k}',t)=g(\vec{k},\vec{k}')h(t)$ where the function $h(t)$ describes the time envelope shape of the polariton pump pulse. The Heisenberg equations of motion for $a_{\vec{k}}$ and $b^{\dagger}_{\vec{k}}$ yield finally to

$$\begin{aligned}\dot{a}_{\vec{k}}(t) &= -i\left(E(\vec{k})-i\Gamma_0+2V_0a_0^{\dagger}a_0\delta_{\vec{k},0}\right)a_{\vec{k}}-i\eta_{\vec{k}}(t)\\ &+ \int_0^t h(t)h(t-\tau)a_{\vec{k}}(t-\tau)K_{\vec{k}}(\tau)d\tau \qquad (3)\end{aligned}$$

where the kernel function is $K_{\vec{k}}(\tau)=\sum_{\vec{k}'}\mid g(\vec{k},\vec{k}')\mid^2 e^{i\omega(\vec{k}')\tau}$ and the exciton noise term is $\eta_{\vec{k}}(\tau)=\sum_{\vec{k}'}g(\vec{k},\vec{k}')b_{\vec{k}'}(0)e^{i\omega(\vec{k}')\tau}h(\tau)$. A phenomenological damping term Γ_0 has been added in Eq.(3) to account for radiation losses from the microcavity. Since the bare energy dispersion relation for the exciton bath in Eq.(1) is simply $\omega(k)=k^2/2M_X$, where M_X represents the exciton effective mass in the quantum well, a Gaussian form for $g(\vec{k},\vec{k}')$ is adequate [8], $g(\vec{k},\vec{k}')=\frac{i\Gamma^{1/2}}{(2\pi\sigma_k^2)^{1/4}}e^{-\frac{(\vec{k}-\vec{k}')^2}{4\sigma_k^2}}$ where Γ settles the polariton-exciton coupling strength maximum and σ_k its width dispersion.

In the following we focus on the time evolution of the mean number of polaritons in the optically active $\vec{k}=0$ state, i.e. $<a_0^{\dagger}a_0>(t)$. We solve numerically Eq.(3). Parameters used correspond to CdTe microcavities. The bottom polariton state corresponds to $E(q=0)=\Omega_0=0.1671606ps^{-1}$ and the exciton mass is $M_X=m_e+m_h=(0.11+0.35)m_0=0.46m_0$. The time evolution of the polariton population in the bottom trap state $\vec{q}=0$ is depicted in Fig.1. After a high intensity pulse excitation the emission at short times starts growing with a nonlinear slope, as has already been shown experimentally [4, 5]. This behavior is a clear manifestation of non-Markovian

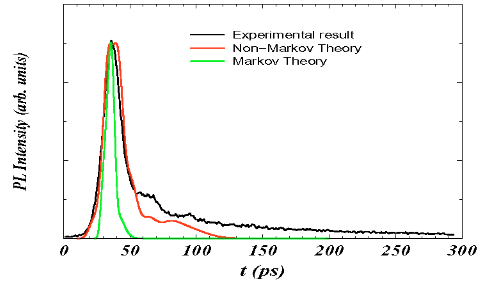

FIGURE 1. Time evolution of the PL: experiment (black line), Non-Markov (red-line)and Markov results (green-line).

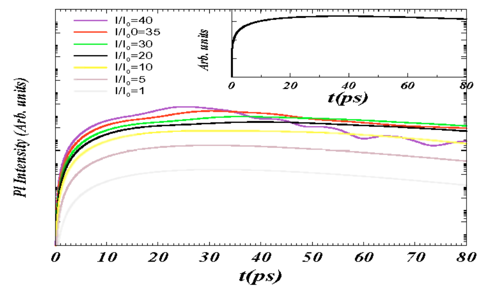

FIGURE 2. Time evolution, on a semi-log scale, of the PL for different pump intensities. The inset shows the pump pulse shape.

effects. For contrast, we also plot the Markov results, i.e. $K(\tau)\sim\delta(\tau)$. Indeed, in the latter case the experimental results cannot be simulated.

In order to show the main differences between the low and high density regimes, we plot in Fig. 2 the results of our model. Clearly, at long times the decays are similar for both high and low intensity pumping, and memory effects are negligible.

In summary, we have demonstrated that observed features in recent polariton PL experiments have their origin in non-Markovian or memory effects, as they are enhanced by a high power excitation. We found that the nonlinear raising and the oscillations/plateaux in the time-resolved polariton PL are well explained by the excitation induced strong coupling between the polariton-exciton systems.

REFERENCES

1. P.G.Savvidis et al., *Semicond.Sci.Technol.* **18**, S311 (2003).
2. J.J.Baumberg et al., *Phys.Stat.Sol.* **(b)242**, 2210 (2005).
3. M.D.Martin et al., *Phys.Rev.Lett.* **89**, 077402 (2002).
4. G.Aichmayr et al., *Semicond.Sci.Technol.* **18**, S368 (2003).
5. L.Viña et al., *Semicond.Sci.Technol.* **19**, S333 (2004).
6. D.Porras et al., *Phys.Rev.* **B67**, 161310 (2003).
7. C.Ciuti et al., *Phys.Rev.* **B62**, 4825 (2000).
8. H.P.Breuer et al., *Phys.Rev.* **A60**, 3188 (1999).