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Detuning dependence of polariton spin dynamics

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Abstract

We have investigated the polariton recombination and spin dynamics in the nonlinear emission regime as a function of the cavity–exciton detuning (δ). The relaxation to $K \sim 0$ states, after non-resonant excitation, is governed by polariton final-state stimulated scattering. The time evolution of the degree of polarization of the photoluminescence ($\wp$) shows a very rich and novel behaviour, never observed in the excitonic emission in bare quantum wells. $\wp$ attains its maximum at a finite time after the excitation with a light pulse instead of monotonically decaying to zero as in the case of bare excitons. Furthermore, the sign of $\wp$ is strongly dependent on the detuning: it is positive for $\delta > 0$ and negative for $\delta < 0$, showing an oscillatory behaviour for $\delta \sim 0$. The change in the sign of $\wp$ with the detuning is correlated with an energy splitting between the σ^+- and σ^--polarized components of the photoluminescence.

The possibility of achieving laser action for very small excitation densities has stimulated an extensive study of the nonlinear emission in semiconductor microcavities in the last few years [1–4]. The development of fast spin-based optoelectronic devices has rekindled the interest in the manipulation of the spin in semiconductors and a new field known as *spintronics* has arisen. The radiation–matter interaction yields the formation of a new quasiparticle, the polariton [5]. In microcavities, there is a considerable enhancement of the light–matter coupling with respect to bare quantum wells (QWs) [6]. Significant differences between the polariton and the exciton spin dynamics are expected due to the exciton–photon mixing and strong anomalies in the polarization of the polariton emission in III–V microcavities have been recently reported [7–9]. In this paper we will give a detailed description of the nonlinear emission and spin polarization dynamics of cavity polaritons in a II–VI microcavity.

The sample is a $\lambda/2$ $Cd_{0.4}Mg_{0.6}Te$ microcavity with two 90 Å thick CdTe quantum wells (QWs) placed at its centre. The top/bottom cavity mirrors are distributed Bragg reflectors made of 17.5/23 pairs of alternating $\lambda/4$-thick layers of $Cd_{0.4}Mg_{0.6}Te$ and $Cd_{0.75}Mn_{0.25}Te$. The strong radiation–matter interaction leads to a Rabi splitting of ~10 meV. The cavity is wedge shaped and allows tuning of the cavity into resonance with the QW exciton by focusing the excitation spot on different points of the wafer. The sample, mounted in a cold finger cryostat where the temperature is kept at 5 K, is non-resonantly excited with σ^+-polarized light pulses (2 ps) at the first minimum above the stop band of the cavity mirrors, approximately 85 meV above the bare cavity mode. The photoluminescence (PL) is time and spectrally resolved using an up-conversion spectrometer. Simultaneously, it is polarization-resolved by analysis into its σ^+- and σ^--polarized components. The degree of circular polarization of the PL is defined as $\wp = (I^{+/+} - I^{+/-})/(I^{+/+} + I^{+/-})$, where $I^{+/-}$ is the intensity of the $\sigma^{+/-}$ component of the PL.

We have studied the recombination and the spin dynamics of microcavity polaritons as a function of the cavity–exciton detuning ($\delta = E_C - E_X$) and the excitation density, ρ_{exc}. The recombination dynamics of the upper/lower polariton (UP/LP) branch is strongly dependent on δ. At low powers, the characteristic decay times (τ_d) of the PL are τ_d (LP) $\sim$ 375/175 ps, τ_d (UP) $\sim$ 15/100 ps for $\delta > 0/\delta \leqslant 0$. Increasing the excitation density drives the system into the nonlinear emission regime. Figure 1(*a*) displays on a semi-logarithmic scale the integrated PL intensity of the LP as a function of ρ_{exc} for $\delta < 0$ at 20 ps, showing an unambiguous exponential

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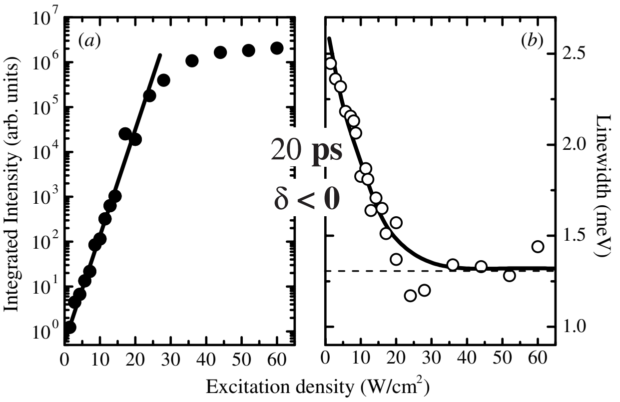

Figure 1. (*a*) Integrated intensity (semi-logarithmic scale) of the LP branch emission at 20 ps as a function of the excitation density for $\delta \sim -10$ meV. (*b*) Linewidth of the LP branch emission at 20 ps as a function of the excitation density for $\delta \sim -10$ meV. The linewidth corresponds to the full width at half maximum extracted from a Gaussian fit of the energy spectrum. The dashed line indicates the experimental energy resolution limit.

dependence. The same behaviour is observed for other detunings. Similar exponential growth has been reported and interpreted in terms of final state stimulated scattering [3], as is characteristic of a bosonic system when the occupancy of the final state approaches unity. Our experimental findings confirm that the dynamics in the nonlinear emission is governed by polariton–polariton stimulated scattering *also* after non-resonant excitation [10, 11]. Figure 1(*b*) depicts the linewidth (full width at half maximum extracted from a Gaussian fit of the energy spectrum) of the LP emission as a function of ρ_{exc} for $\delta < 0$ at 20 ps. A reduction of a factor 2 is already seen at 20 W cm^{-2}, limited by the spectral resolution of the experimental setup. Recently, a reduction of approximately two orders of magnitude in the polariton emission linewidth with the excitation power has been theoretically predicted and interpreted as a characteristic feature of polariton lasing [12]. In the following we will consider only the nonlinear emission regime and concentrate on the spin dynamics of cavity polaritons for different characteristic detunings.

Negative detuning

For this detuning the LP has a predominantly photonic character, the dispersion relation around $K \sim 0$ is strongly distorted due to the very light mass of the polariton and the density of states is reduced. At low powers, when a small number of polaritons are excited in the system, the relaxation towards $K \sim 0$ is hindered by a bottleneck in the LP branch at large K [13, 14]. However, for larger excitation powers, when the system is in the nonlinear regime, the polaritons relax very efficiently from bottleneck states to $K \sim 0$ through final-state stimulated scattering. Our experimental results confirm the very efficient relaxation to $K \sim 0$ and demonstrate that the recombination process is also accelerated in the nonlinear regime as the majority of the emission occurs within the first 75 ps after excitation. The spin dynamics for $\delta < 0$ is also accelerated. Figure 2 depicts the time evolution of the polarization for $\rho_{\text{exc}} = 60$ W cm^{-2}. $\wp$ is positive at $t = 0$, mirroring the spin population imbalance created by the non-resonant excitation, i.e. 2/3 heavy holes (involved in the formation of +1 spin excitons) and 1/3 light holes (to form –1 spin excitons). Shortly after, the polarization switches to negative values as a result of a predominant counter polarized emission, reaching its maximum value ($\sim$–75%) in a very short time (this time depending strongly on ρ_{exc}, shortening with increasing excitation). After reaching this maximum, $\wp$ decreases very quickly to zero, following the very fast dynamics of the emission, and the co- and counter-polarized emission intensities remain equal. The excitation pulse initially creates a larger +1 spin population but on relaxation to $K \sim 0$ states, the stimulated scattering is more efficient to –1 spin states than to +1. The accumulation of –1 spin polaritons results in a larger σ^- polarized emission and therefore a large negative polarization. This negative $\wp$ cannot be attributed to resonant excitation of the light hole exciton transition as the excitation energy is more than 30 meV above it. The different scattering efficiencies could be related to an energy splitting ($\Delta = E^- - E^+$, where $E^{+/-}$ denotes the emission energy of the $\sigma^{+/-}$ polarized emission) observed between the +1 and the -1 spin levels at $K \sim 0$. This splitting is depicted in the inset of figure 2. Δ increases with excitation density, saturating at $\sim$–0.6 meV for $\rho_{\text{exc}} > 40$ W cm^{-2}. This splitting is qualitatively similar to that found in bare QWs [15, 16]. Thus, even though in the case of bare QWs the majority of the emission was co-polarized with the excitation, the exciton–exciton interaction seems to be involved in the occurrence of the spin splitting of the $K \sim 0$ states of the LP branch.

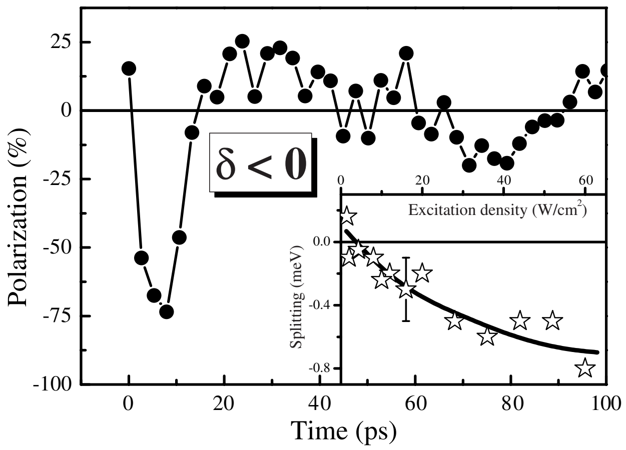

Figure 2. Time evolution of the circular polarization degree of the PL emission from the LP at $\delta \sim -10$ meV for an excitation density $\rho_{\text{exc}} = 60$ W cm^{-2}. Inset: spin splitting at 20 ps as a function of the excitation density. The line is a guide to the eye.

Positive detuning

For this detuning, the LP has a predominant excitonic character. The nonlinear PL spectrum shows three peaks, which correspond to the emission from the LP branch, the bare cavity mode and the UP branch, respectively. The coexistence of coupled and uncoupled modes in a microcavity has already been reported in the literature [17, 18]. The bigger nonlinearity is observed in the emission from the bare cavity

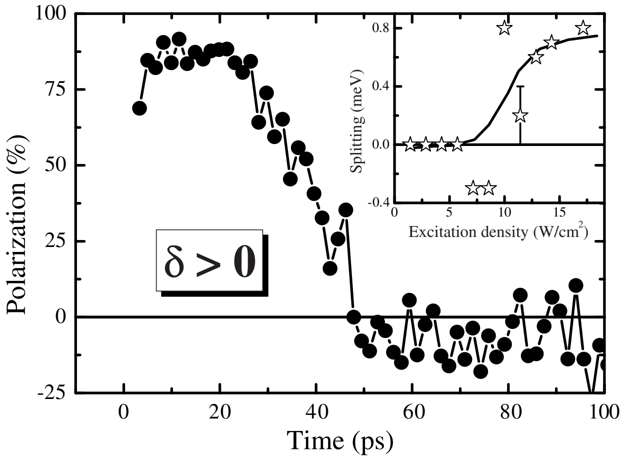

Figure 3. Time evolution of the circular polarization degree of the PL emission from the cavity mode at $\delta \sim 10$ meV for an excitation density $\rho_{\mathrm{exc}} = 20$ W cm^{-2}. Inset: spin splitting at 20 ps as a function of the excitation density. The line is a guide to the eye.

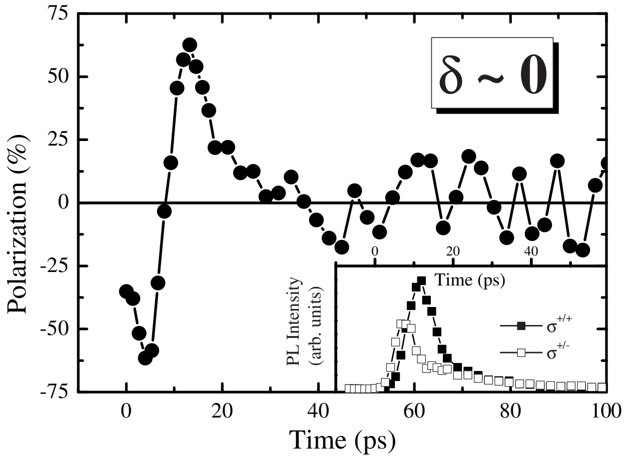

Figure 4. Time evolution of the circular polarization degree of the PL emission from the LP at $\delta \sim 0$ meV for an excitation density $\rho_{\mathrm{exc}} = 60$ W cm^{-2}. Inset: σ^+ (solid squares) and σ^- (open squares) polarized time evolution traces for $\delta \sim 0$ meV and $\rho_{\mathrm{exc}} =$ 60 W cm^{-2}.

mode. The spin dynamics is remarkably different from the case of negative detuning, as now the majority of the emission is co-polarized with the excitation. Figure 3 shows the polarization degree of the emission as a function of time for $\rho_{\mathrm{exc}} \sim$ 20 W cm^{-2}. Instead of monotonically decaying to zero as in the case of bare QWs or reaching negative values as in the case of $\delta < 0$, $\wp$ increases from its initial value up to almost 100% at ~20 ps delay. After reaching its maximum, the polarization decays to zero as the intensities of both circularly polarized components of the emission become equal. After the initial excitation with a σ^+ polarized pulse, the scattering to $K \sim 0$ states is more efficient now to the +1 spin level and it results in an almost completely co-polarized emission. As in the case of negative detuning, we have observed an energy splitting between the +1 and the -1 spin states, which is depicted in the inset of figure 3. In contrast to the case of $\delta < 0$, it is positive for $\delta > 0$, i.e. the σ^+ polarized emission lies at lower energies than the σ^-. There is no splitting for small ρ_{exc} but it increases with excitation density, saturating at ~0.6 meV for $\rho_{\mathrm{exc}} \sim$ 20 W cm^{-2}. The fact that the splitting is now positive disagrees with the exciton–exciton interaction argument described in the previous section for $\delta < 0$. The result of such interaction will be the majority population (+1 spin) at higher energies than the minority (-1 spin), leading to a negative splitting, in total opposition to our experimental findings for $\delta > 0$. Still it seems that the many-body interaction between the excitonic parts of the polaritons is somehow involved in the development of the spin splitting.

Resonance

In this section we will focus on the description of the polariton spin dynamics for the case in which the exciton and the cavity modes are in resonance ($\delta \sim 0$). The polariton is half exciton/half photon under these conditions. The spin dynamics in this case is even more striking than those described in the previous two sections. The degree of polarization of the emission of the LP is depicted in figure 4 for an excitation density of 60 W cm^{-2}. A very fast oscillation in $\wp$ can be observed, changing from -60% to +60% in ~10 ps. A similar oscillatory behaviour has been reported in a slightly positively detuned GaAs microcavity [7], although the oscillation was much slower and had a π phase shift. Such a time evolution of the polarization implies that the emission is initially counter (σ^-) polarized and then very quickly becomes co (σ^+) polarized, decreasing then to zero. The inset in figure 4 depicts the polarization-resolved time evolution of the LP emission for $\rho_{\mathrm{exc}} \sim$ 60 W cm^{-2}. It shows that the σ^- polarized emission peaks earlier than the σ^+. Very recently a theoretical model has been developed by Kavokin and co-workers [19], within the pseudospin framework, which accounts for the oscillations of $\wp$. In this model, the mechanism responsible for the oscillatory behaviour is the TE/TM splitting of the exciton or equivalently, the intrinsic birefringence of the cavity. A detailed study of the LP emission spectra is still underway. Preliminary results show that there is also an energy splitting (Δ) between the two circularly polarized components of the PL: Δ is negative for low excitation densities and then it decreases with ρ_{exc}, eventually reaching positive values for larger excitation densities. However, an exhaustive description of the spin splitting dependence on the excitation density for the case of $\delta \sim 0$ will be given elsewhere [18].

In summary, we have presented the recombination and the spin dynamics of polaritons in the nonlinear emission regime after non-resonant excitation. We have shown that final state stimulated scattering is the main mechanism governing the energy and momentum relaxation process, as made evident by the exponential rise of the emission intensity with excitation power. We have observed that the nonlinear emission from the photon-like branch is strongly polarized, either parallel ($\wp \sim$ 100% for $\delta > 0$) or perpendicular ($\wp \sim -80$% for $\delta < 0$) to the excitation, showing a marked oscillatory behaviour for $\delta \sim 0$. We have found an energy splitting between the two circularly polarized components of the emission which is most likely related to the interaction between the excitonic parts of the polaritons. The spin splitting is positive/negative for

$\delta > 0/\delta < 0$ and reveals that there is a reversal in the spin alignment of $K \sim 0$ states with the exciton–cavity detuning.

Acknowledgments

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