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Polariton Spin Dynamics in II–VI Microcavities

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The spin dynamics of cavity polaritons, in the non-linear regime, present a novel and striking behaviour of the degree of polarization of photoluminescence. It reaches its maximum value at a finite time after excitation with a light pulse, and is strongly influenced by the exciton–cavity detuning. In the case of negative detuning, the relaxation towards $K \sim 0$ states is governed by the polariton final state stimulated scattering, the emission is counter-polarized with the excitation and the polarization reaches very large negative values. In the case of positive detuning, the emission arises from the bare cavity mode, indicating a transition to the weak coupling regime; nevertheless, a very large positive polarization is observed. In addition to the large degrees of polarization of the emission, an energy splitting is observed between the two circularly polarized components of the photoluminescence, which is directly related to the anomalous behaviour of the polarization.

Introduction Semiconductor microcavities have been widely studied in the last years since the first experimental observation of strong exciton–cavity coupling [1]. The possibility of achieving laser action for very small excitation densities has stimulated the exhaustive study of the properties of the non-linear emission in these systems, and many experimental reports concerning both III–V [2–4] and II–VI [5–7] microcavities have been published. Simultaneously, there has been a renewed interest in the manipulation of the spin in semiconductor heterostructures. In the particular case of microcavities, strong polarization anomalies have been reported recently, indicating the significant role played by the spin in the dynamics of cavity polaritons [8–11]. In the present paper we give a detailed description of the spin polarization dynamics of cavity polaritons in II–VI microcavities in the non-linear emission regime.

The sample under study is a $Cd_{0.40}Mg_{0.60}Te$ microcavity of thickness $\lambda/2$, sandwiched between the top (bottom) distributed Bragg reflectors (DBRs). These mirrors are made of 17.5 (23) pairs of alternating $\lambda/4$ thick layers of $Cd_{0.40}Mg_{0.60}Te/Cd_{0.75}Mn_{0.25}Te$. Two 90 Å thick CdTe quantum wells are placed in the centre of the cavity to obtain the optimum radiation–matter interaction, which leads to a Rabi splitting of ~10 meV. A slight wedge in the cavity thickness allows tuning of the cavity and the exciton into resonance by moving the excitation spot across the wafer. The sample is mounted in a cold-finger cryostat where its temperature is kept at 5 K and is optically excited at the first minimum above the stop-band of the DBRs. The emitted photoluminescence (PL)

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is time and spectrally resolved using an up-conversion spectrometer with a time resolution of ~2 ps. For polarization-resolved measurements, a pair of $\lambda/4$ plates are included in the experiment: the excitation light is σ^+ polarized and the PL is resolved into its σ^+- and σ^--polarized components.

We have studied the time evolution of the polariton PL as a function of the cavity–exciton detuning ($\delta = E_C - E_X$) and as a function of the excitation power density. The recombination dynamics of the upper/lower polariton (UP/LP) branch are strongly dependent on the detuning. At low powers, the characteristic decay times are $\tau_d(LP) \sim 375/175$ ps and $\tau_d(UP) \sim 15/100$ ps for $\delta > 0/\delta \leq 0$, respectively. Increasing the excitation density drives the system to the non-linear emission regime. We have observed that the stimulated emission originates from the polariton branch with the largest photonic content, i.e. the LP/UP for $\delta < 0/\delta > 0$. The integrated emission intensity of this photon-like polariton branch displays an exponential growth with excitation power. Similar exponential growths have been reported in the literature and have been attributed to bosonic final state stimulated scattering [3, 9].

In the case of negative detuning, at high levels of excitation the interaction between excitons and cavity photons remains in the strong coupling regime and, therefore, the non-linear emission is due to polariton stimulation. On the contrary, in the case of positive detuning, the system has transited to the weak coupling regime from the smallest excitation powers, as evidenced by the energy shifts of the polariton branches to the bare exciton/cavity mode. In this case, the emission is due to photon stimulation, as in a conventional laser. In the case of zero detuning, an intermediate behaviour is displayed: at low excitation powers excitons and photons are strongly coupled, but a transition to the weak coupling regime is observed for moderate excitation powers.

In this paper we concentrate on the description of the polariton polarization dynamics in the non-linear regime for the cases of negative (strong coupling) and positive (weak coupling) exciton–cavity detuning.

Negative Detuning Negative detuning is characterized by a LP with a large photonic content. The polariton dispersion relation around $K \sim 0$ is distorted by the very small mass of the photon mode and a bottleneck in the relaxation to these states is observed at low powers [12, 13]. Our results reveal that the non-linear PL peaks at very short times (~20 ps). Figure 1 depicts the time evolution of the degree of polarization of the LP's PL for an excitation density of ~20 W/cm^2. The initial degree of polarization is ~40% but it changes very quickly to –75% at ~20 ps. After reaching this minimum, the polarization returns to zero. The initial σ^+-polarized pulse creates a larger +1 spin population, which is reflected at $K \sim 0$ by the positive degree of polarization at $t = 0$. The relaxation of the non-resonantly created polaritons to $K \sim 0$ is governed by the final state stimulated scattering. Nevertheless, the scattering to the –1 spin states is more efficient than to the +1 spin states. The accumulation of –1 spin polaritons results in a larger σ^--polarized emission and, therefore, a very large negative polarization. This negative polarization could be attributed to a resonant excitation of light-hole excitons; however, this possibility can be discarded by energy arguments (the excitation energy is always at least 30 meV above the light-hole exciton resonance) and, furthermore, such a resonant excitation would lead to a negative initial degree of polarization, in contrast with the experimental findings.

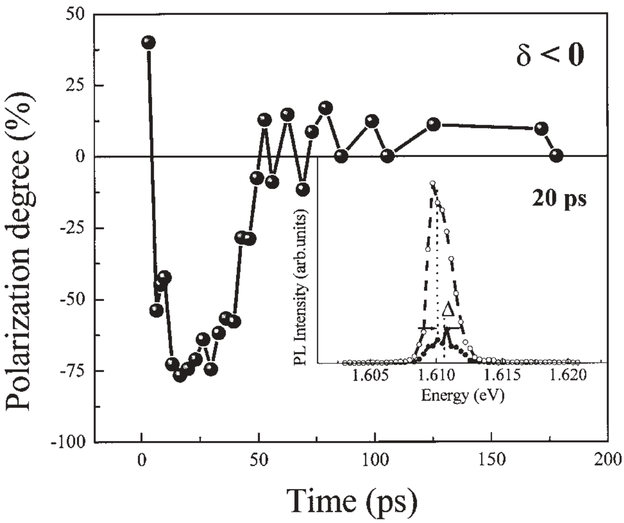

Fig. 1. Time evolution of the degree of polarization of the emission of the LP branch for an excitation density of 20 W/cm^2 and a detuning of −10 meV. The inset shows the polarization-resolved PL spectra (σ^+, solid line; σ^-, dashed line) for a delay time of 20 ps, $\delta = -10$ meV and an excitation power of 20 W/cm^2

The different scattering efficiencies might be related to an energy splitting observed between the two circularly polarized components of the PL at very short times. This splitting mirrors the different energies of the +1 and the −1 spin levels and is depicted in the inset of Fig. 1 for a delay time of 20 ps (which is the time when the emission reaches its maximum intensity) and an excitation density of 20 W/cm^2. This spin splitting increases with excitation power, saturating at ∼0.5 meV for 20 W/cm^2. As can be deduced from the inset of Fig. 1, the energy of the −1 spin states at $K \sim 0$ is less than that of the +1 spin states. This fact, in addition to the different efficiencies of the +1/−1 stimulated scattering processes, would account for the large σ^--polarized emission intensity and the observed negative polarization. Therefore, the increase of the negative polarization ($0 < t < 20$ ps) is due to the polariton stimulated scattering to the state of lowest energy at $K \sim 0$ (i.e. the −1 spin state). The accumulation of −1 spin polaritons disappears through the stimulated emission process, eventually equalizing the +1 spin population, taking the polarization to zero very quickly (20 ps $< t <$ 50 ps). However, this is only a qualitative interpretation of the experimental results and further studies are underway to characterize in detail this polarization reversal and the origin of the spin splitting.

Positive Detuning As mentioned above, for positive exciton–cavity detuning, the system has transited to the weak coupling regime. Already at relatively small excitation powers the emission energies of both polariton branches have already shifted to those of the bare exciton (LP) and photon (UP) modes. Therefore, the non-linear emission observed is due to conventional photon stimulation. However, very interesting polarization effects are still observed.

Figure 2 depicts the time evolution of the degree of polarization of the cavity-like mode PL for an excitation density of 20 W/cm^2. The initial degree of polarization is ∼40% and it increases up to a maximum value of 90% at 20 ps, after which the polarization decreases to zero. The increase of the degree of polarization ($0 < t < 20$ ps) can be interpreted as follows. The initial σ^+-polarized pulse creates a larger +1 spin popula-

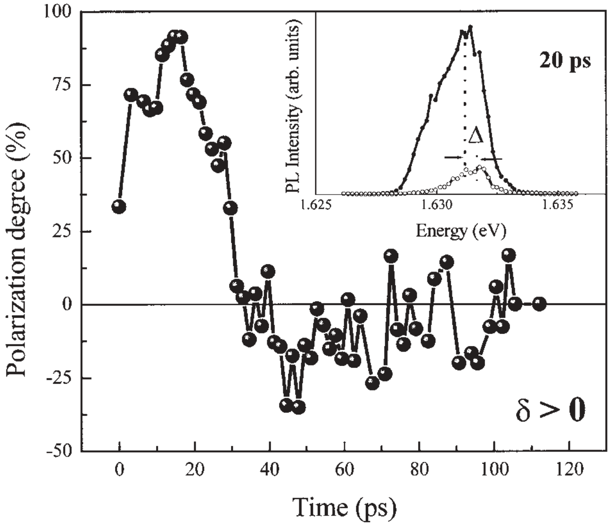

Fig. 2. Time evolution of the degree of polarization of the emission of the cavity mode (UP branch) for an excitation density of 20 W/cm^2 and a detuning of +10 meV. The inset shows the polarization-resolved PL spectra (σ^+, solid line; σ^-, dashed line) for a delay time of 20 ps, $\delta = +10$ meV and an excitation power of 20 W/cm^2

tion. A fast scattering process will bring all the photo-created excitons to the cavity mode before any spin relaxation can occur, which will result in a larger +1 spin population of $K \sim 0$ states at $t = 0$. This initial +1 spin population will act as a seed for the stimulated scattering process and therefore a large number of +1 spins will be transferred to $K \sim 0$ states. The radiative recombination of this population will result in a bigger σ^+-polarized emission, i.e. a very large positive polarization. After reaching the maximum +1/−1 spin population difference ($t \sim 20$ ps), the +1 spin population disappears very quickly through the σ^+-polarized stimulated emission process, taking the polarization to zero ($20 < t < 40$ ps).

The polarization-resolved PL plot (inset of Fig. 2) reveals an energy splitting of the two circularly polarized components, similar to that observed for negative detuning. However, in this case, it is the σ^+-polarized component that lies at lower energy, indicating that for $\delta > 0$ it is the +1 spin state that is at lower energies at $K \sim 0$. This splitting increases with excitation density, saturating at ~0.8 meV for 20 W/cm^2. As in the case of negative detuning, the bosonic stimulated process transfers most of the bosons to the spin level of lowest energy at $K \sim 0$, which results in a larger σ^+-polarized emission.

Conclusions It has been shown that bosonic stimulated scattering is the main relaxation mechanism for non-resonantly created cavity polaritons in the non-linear emission regime. We have observed very large degrees of circular polarization of the emission, which are related to the lifting of the spin degeneracy at $K \sim 0$ at very short times. The spin of the lowest energy level changes from +1 to −1 on changing the exciton–cavity detuning from positive to negative. The physical origin of this reversal of the spin alignment is still under study.

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