Physics of Semiconductors 2002. Institute of Physics Conference Series 171, P106 (2003), ed. by A R Long and J H Davies (Institute of Physics Publishing, Bristol, UK)

Spin dynamics of polaritons in II-VI microcavities: detuning dependence

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Abstract. We have studied the polarization of the light emitted by a semiconductor microcavity as a function of the detuning between the cavity-mode and the exciton. The time dependence of the polarization, which represents the spin dynamics of the polaritons, shows a very rich and novel behaviour in this non-linear regime, as compared to that under low excitation conditions. The sign of the polarization is strongly dependent on the cavity-exciton detuning ($\delta = E_C - E_X$): it is positive for $\delta > 0$ and negative for $\delta < 0$. The negative polarization is directly related with an energy splitting between the σ^+- and σ^--polarised components of the emission, which appears when the excitation density drives the cavity into the non-linear regime.

1. Introduction

Since the so called strong coupling between exciton and cavity photon to cavity polaritons was first demonstrated by Weisbuch *et al*. [1], the field has attracted immense interest in solid state physics. Apart from other new phenomena it was soon realized that bosonic effects, which in theory should be observable in bare quantum wells (QWs) but were not, could become a reality in microcavities. Whereas in the former case the required particle densities to access boson statistics were too high, in the latter systems however, this is possible due to an altered density of states (DOS) in the strongly coupled polariton system. Like excitons, polaritons can be described as Bosons, due to their integer spin, provided that their densities are sufficiently low so that interaction between the particles is small [2- 4]. Whether this approximation is valid or not, can be obtained from an inspection of the commutator of the Boson operators, which equals approximately $1 - 2\pi a_B^2 n_X$ [5]. Thus the deviation of the commutator from unity is directly proportional to the exciton density, n_X, and to the square of the Bohr radius, a_B, i.e. the bosonic approximation is only valid, if $2\pi a_B^2 n_X \quad 1$. Kira *et al*. calculated the Bose commutator for GaAs QWs that were incorporated into a microcavity and found that it is only close to 1 when $n_X \leq 10^{10}\text{cm}^{-2}$ [6]. Since the Bohr radius in GaAs QWs is about 150 Å and in CdTe ~30 Å, the Bose limit is valid to an exciton density at least one order of magnitude higher in the CdTe system than in the GaAs case. In CdTe systems, which will be studied here, the bosonic approximation is valid up to densities of $n_X \approx$

$10^{11}cm^{-2}$. When Bose statistics is valid, one state can be occupied by more than one particle. It can be shown that when the occupation number of such a state reaches one, the rate at which particles relax from the excited state to the ground state increases exponentially. Therefore this process is called final state stimulation of the polariton relaxation. Relaxation processes are usually mediated by phonon emission, but it has been predicted a few years ago [7], that also direct polariton-polariton scattering can lead to final state stimulation.

Recently two experimental works were published almost simultaneously, and independently from each other, which indicate final-state-stimulation relaxation in microcavities at zero and negative detunings [8,9]. In the pump and probe experiments of Ref. 9, an exponential increase of the gain with increasing excitation density was obtained in an InGaAs microcavity, indicating final state stimulation. The onset of the positive gain was accompanied be a characteristic blue shift of ~ 1 meV, which then remained constant on further increasing the intensity. This shift seemed to be intrinsic to the process and did not correspond to the breakdown of the strong coupling in the system, since densities were well below saturation. A follow-up experiment by the same group yielded the angle-resolved and time integrated emission from the same system [10]. Ciuti *et al*. could reproduce the experimental features by calculations done in the polariton basis assuming their bosonic character [11,12]. Similar results were achieved by continuous wave pumping of the lower polariton branch at a certain angle [13,14].

The subject of the coexistence of non-linear effects and the strong coupling regime under non-resonant excitation has been a matter of debate. Some authors claim that these effects coexist in InGaAs/GaAs microcavities [15,16], at densities where the Bose commutator still is close to one. While others claim that in III-V microcavities stimulated polariton scattering cannot be achieved under high-energy non-resonant excitation conditions [17]. However, in II-VI microcavities, and in particular in CdTe based materials, the existence of stimulated scattering and probe amplification under non-resonant conditions seems to be well established [18-20].

Only very recently investigations of the spin dynamics in microcavities have been performed, in spite of the new possibilities that the strong modification of light-matter interactions offer to manipulate the spin degree of freedom. The exponential increase of the gain [9] is highly spin dependent and occurred only for co-circular polarizations of the pump and probe beams. The spin-dependence of the gain is indeed more complicated and it has been also shown that for a circularly-polarized probe the maximum gain is obtained with a linearly-polarized pump pulse [21]. Long spin-relaxation times at zero detuning (δ=0) and stimulated scattering into spin-polarized states at the bottom of the lower polariton branch have been reported on cw [15] and time-resolved [22] photoluminescence experiments. In GaAs microcavities strong non-linear emission was observed and an oscillatory behaviour of the circularly polarized emission in the time domain was reported [23]. The occurrence of a negative polarization of the non-linear light emission in the strong coupling regime of a CdTe microcavity has been reported very recently in the literature [24]. The angular dependence of this phenomenon is presented in another contribution in these Proceedings [25]; in the present work this system is studied in detail in its polarization dynamics as a function of cavity- to exciton-mode detuning and excitation intensity.

2. Experimental details

The sample under study is a $\lambda/2$ $Cd_{0.4}M_{0.6}Te$ wedge-shaped cavity. In the antinode position of the electromagnetic standing wave, at the centre of the cavity, are

placed two CdTe QWs of 90 Å. The top/bottom cavity mirrors are Distributed Bragg Reflectors (DBRs) made of 17.5/23 pairs of alternating layers of $Cd_{0.4}Mg_{0.6}Te$ and $Cd_{0.75}Mn_{0.25}Te$. The sample was held at 5 K in a cold finger cryostat. In order to obtain uniform excitation conditions for all detunings, δ, the same non-resonant excitation conditions were chosen, with the excitation energy, E_{exc}, tuned to the first reflectivity minimum following the stop-band of the DBRs. This minimum lays 84 meV above the bare cavity mode, regardless of detuning position. The time-resolved photoluminescence (tr-PL) measurements were done in a standard one-colour up-conversion set-up with 2 ps pulses provided by a Ti:Spphire laser. The excitation pulses were circularly σ^+-polarized. Due to the excitation conditions with ΔE_{exc}=84 meV, the accessible cavity detuning was limited to δ=15.6 meV by the available maximum energy of the laser. The excitation densities, I_{exc}, are obtained, assuming an excitation spot diameter of 100 μm, a laser repetition frequency of 82 MHz and a pulse width of 2 ps. With an absorption of ~1% at these excitation energies [19] and with I_{exc}=1 Wcm^{-2}, an exciton density of $n_X \approx 5x10^8 cm^{-2}$ is created. The emission from the microcavity was detected in a solid angle of ± 2º around the sample normal, i.e. only polaritons with $\mathbf{K} \leq 0.3x10^4 cm^{-1}$ are up-converted and measured. In most cases, the upper polariton (UP) emission was too faint to be detected with sufficient signal to noise ratio. Therefore the characterization of the dynamics is focused on the lower polariton (LP) branch emission.

3. Results and discussion

The time evolution of the emission is presented in Fig. 1 for different detunings, where solid (dashed) lines show the co-(counter-) polarized components. The left panels correspond to the LP branch under low excitation conditions: an increase in the decay rate is clearly seen going from positive (δ= +15 meV) to negative (δ= -21 meV) detunings, while the difference in intensity of both polarized components is very small and independent of δ. This behavior is quite similar to that obtained for the PL of excitons in bare quantum wells.

However, this situation changes drastically when the system is brought into the non-linear emission regime, as can be observed in the right panels of Fig. 1, which correspond to an excitation power of 15 mW, and the emission stems from the branch with the largest photonic content. In first place all the dynamics of the polaritonic light emission becomes much faster under high excitation conditions, what already hints the presence of stimulation processes [23] (note that the scales of the left/right panels span up to 600 ps/150 ps). For δ= +15 meV the co-polarized emission ($\sigma^+\sigma^+$) is considerably larger than the counter-polarized ($\sigma^+\sigma^-$). The difference becomes even larger and the decay rate of the stimulated component of the emission (σ^+) also increase at δ= +8 meV. However, the intensities become comparable close to resonance (δ= 0 meV) and the counter-polarized emission becomes dominant for negative detunings. It is also worthwhile mentioning that the integrated emission intensity of this photon-like polariton branch displays an exponential growth with the excitation power. Similar exponential growths have been reported in the literature and have been attributed to bosonic final state stimulated scattering [9,13,18].

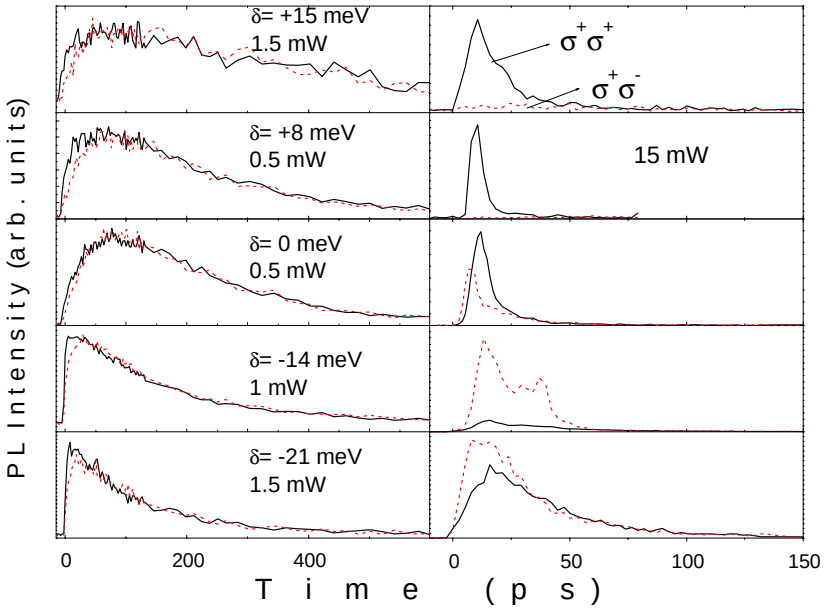

Figure 1. Time dependence of the CdTe/$Cd_{0.4}Mg_{0.6}Te$ microcavity emission for different detunings. The left (**right**) panels correspond to conditions of weak (**strong**) excitation. Solid (dashed) lines represent co- (counter-) polarized circular emission with respect to that of the exciting pulses. Note that at negative detunings, under strong excitation, the σ^- emission is larger than the σ^+-polarized one.

For negative detunings, the difference between the σ^- and σ^+ intensities becomes the largest at $\delta \approx$ -14 meV, where a very peculiar dynamics is obtained exhibiting oscillations in the emitted intensities. This behaviour is discussed in more detail in Ref. 25. A closer look to the initial curvature of the time traces, under high excitation conditions, reveals that those with larger intensity (i.e. σ^-/σ^+ for $\delta<0/\delta>0$) have a changed sign (positive) with respect to those of the traces at low excitation (negative). When the carrier relaxation is mediated by a constant rate, a rise with negative curvature is expected [26]. A positive curvature indicates a relaxation mechanism that is based on an increasing rate, like final state stimulation.

The time evolution of the degree of polarization, defined as $\tilde{A} = \frac{I^{++} - I^{+-}}{I^{++} + I^{+-}}$, with I^{++} (I^{+-}) the emission intensity exciting with σ^+ polarization and detecting the $\sigma^+(\sigma^-)$ component, is depicted in Figure 2. The open points correspond to the low excitation regime, which shows a dynamic of the degree of polarization independent of the detuning and very similar to that found for bare excitons in quantum wells. The traces with the full symbols are taken at an excitation density of 15 mW and demonstrate the very reach behavior of the degree of polarization when the cavity is in the non-linear emission regime. We will concentrate from now on only on the high excitation regime.

At positive detunings, the degree of polarization is positive and it increases up to its maximum value, which can be as high as 90%, in 20 ps; after that, the polarization decreases to zero. The rise of the polarization degree ($0 < t < 20$ ps) can be interpreted as follows: the initial σ^+-polarized pulse creates a larger +1 spin population. A fast scattering process will bring all the photocreated excitons to the cavity mode before any

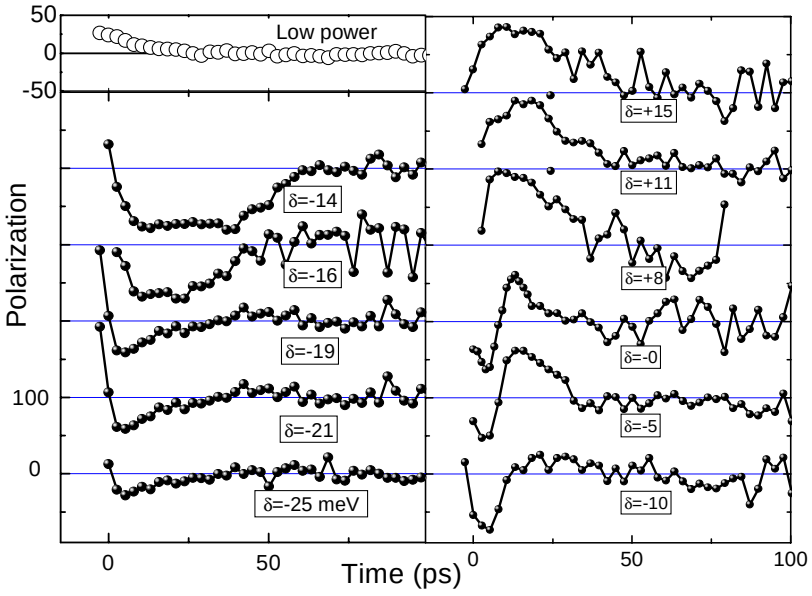

Figure 2. The open circles show the polarization for weak excitation conditions (practically independent of the detuning). Solid circles correspond to the polarization in the non-linear regime (15 mW excitation power) for different detunings. The polarization reaches very large values and changes from negative to positive close to resonance (0 detuning).

spin relaxation can occur, what will result in a larger +1 spin population of **K** ~ 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 ~ 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 ~ 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). This initial rise of the polarization is unique to microcavities and contrasts with the behavior of $\wp$ for bare excitons that always decays with time monotonically to zero from its maximum initial value. Oscillations of the polarization can be hinted in several traces shown in Fig. 2, this behavior is described in more detail in Ref. 25.

When δ is close to zero, the degree of polarization presents a very short time-interval of negative values, followed by a maximum, before $\wp$ decreases steadily to zero. At negative detunings, $\wp$ becomes basically negative reaching values as large as –90% for δ ~ -10 meV. It even shows a plateau lasting ~ 30 ps at δ = -14 meV, before the absolute values of $\wp$ begin to decrease when the detuning is further increased to negative values. This behavior is related to the spin-selective polariton-polariton stimulated scattering to the final state, which is strongly dependent of the exciton-cavity detuning.

In the case of δ < 0, the initial σ^+-polarized pulse creates a larger +1 spin population, which is reflected at **K** ~ 0 by the positive polarization degree at t = 0. The relaxation of the non-resonantly created polaritons to **K** ~ 0 is governed by the final state stimulated scattering. Nevertheless, the scattering to the –1 spin states is more efficient than to +1 spin states. The accumulation of –1 spin polaritons results in a larger σ^--polarized emission and therefore in 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 polarization degree, in contrast with the experimental findings. One could think that the observation of negative polarization contradicts the conservation of angular momentum, but this is not the case, as shown in Ref. 25, because the emission at large angles of observation has positive polarization.

The different scattering efficiencies might be related with an energy splitting observed between the two circularly polarized components of the PL at very short times. This splitting is mirroring the different energies of the +1 and the –1 spin levels and is demonstrated in Figures 3a and 3b, for a delay time of 20 ps (which is the time when the emission reaches its maximum intensity) and an excitation power of 15 mW, for negative and positive detunings, respectively. In the case of $\delta>0$, the splitting is observed only for the UP branch. At negative (positive) detunings, the energy of the –1 (+1) spin states at $\mathbf{K} \sim 0$ is smaller than that of the +1 (–1) spin states; this fact, in addition with the different efficiencies of the +1/-1 stimulated scattering processes would account for the large σ^- (σ^+)-polarized emission intensity and the observed negative (positive) polarization. The fact that the splitting increases with excitation power density indicates that it could originate from exciton-exciton interactions. A theory for bare excitons would qualitatively explain the splitting and the ±1 level ordering, as a result of exchange and vertex corrections to the self-energies, but only for $\Delta<0$ [5].

Let us now look into the situation of negative detunings in more detail. Figure 4 compiles the main features observed for a representative detuning of δ= -10 meV. In panel (a) the dependence of the degree of polarization on excitation power is shown together with the intensity values of the time integrated PL. Up to an excitation power of 4 mW, the σ^+ and σ^- emissions, which rise exponentially, have the same value and thus

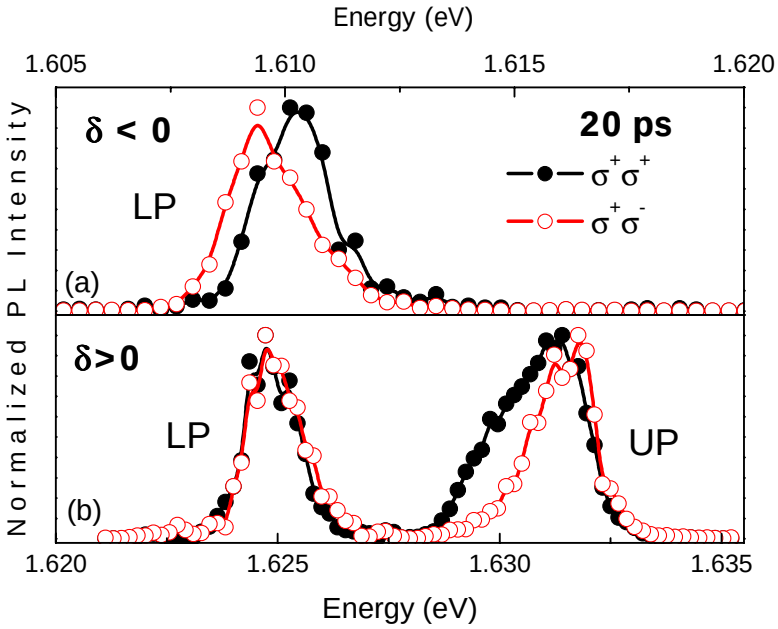

Figure 3. Spectra of the CdTe microcavity taken 20 ps after excitation with a σ^+-polarized pulse analyzed into its σ^+ (●) and σ^- (○) components for an excitation power of 15 mW. Panel (a) corresponds to a negative detuning of –10 meV, while (b) depicts the results for δ = + 10 meV.

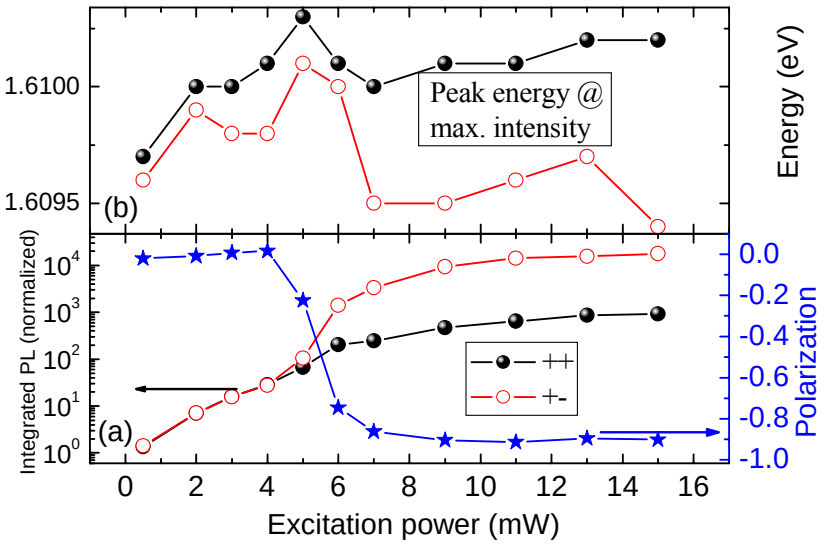

Figure 4. (a) Integrated emission as a function of excitation power for σ^+ (σ^-) polarization, shown with solid (open) circles, at a detuning of –10 meV. The starts represent the degree of polarization (right scale from 0 to –1). (b) Energy of the emission peak at its maximum intensity as a function of excitation power.

the polarization vanishes. For larger excitation intensities, the negative-polarized component becomes larger than the positive one, leading to a sharp drop in $\wp$ to negative values. At 8 mW the maximum negative polarization of $\wp$= –0.9 is reached, which saturates at this value for higher excitation densities. Also the increase of the emission intensity starts to saturate at this excitation value. The bosonic final state stimulation mechanism is therefore the underlying process responsible for the observed dynamics. Figure 4b shows the peak energies as a function excitation power at the time delay when the maximum emission intensity is reached. After an initial blue shift of ~0.5 meV for both components, a splitting between the two orthogonal polarized components of the emission appears. The splitting starts simultaneously with the sign change of the polarization and the enhancement of the σ^--emission as compared to that of the σ^+-polarized one. The behaviour depicted in Fig. 4b for the energy positions of the emission, for powers larger than 4 mW, is qualitatively similar to the one found for bare QWs [27,28], although here the splitting is a factor of ~5 smaller.

5. Summary

In the non-linear regime, the spin dynamics of the photon-like polaritons is strongly dependent on the exciton-cavity detuning. For $\delta > 0$, the UP emission is always co-polarised with the excitation, with the maximum value of the polarization attained at a finite time, ~ 20 ps, reaching almost 100%. At resonance, $\delta = 0$, the polarization presents a sharp negative peak, at very short times, followed by a positive maximum before it decays to zero. The polarization of the LP becomes negative for $\delta < 0$, reaches values as large as –90%, and exhibits a plateau lasting 30 ps at δ = -14 meV. The different behaviour for positive and negative detunings is linked to an energy splitting between the +1 and the –1 spin states at K ~ 0 in the photon-like polariton branch. For $\delta < 0$, the appearance of the splitting, as the excitation density is increased, occurs simultaneously with the reversal of sign of the polarization and with a non-linear increase of the σ^--polarised emission, which surpasses that of the σ^+-emission by almost two orders of magnitude.

Acknowledgments

This work has been partially supported by the Spanish DGICYT (PB96-0085) and the CAM (07N/0064/2001). We thank D. Porras and C. Tejedor for helpful discussions.

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