

Polarization Control of the Nonlinear Emission of Semiconductor Microcavities

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The degree of circular polarization (\wp) of the nonlinear emission in semiconductor microcavities is controlled by changing the exciton-cavity detuning. The polariton relaxation towards $\mathbf{K} \sim 0$ cavitylike states is governed by final-state stimulated scattering. The helicity of the emission is selected due to the lifting of the degeneracy of the ± 1 spin levels at $\mathbf{K} \sim 0$. At short times after a pulsed excitation \wp reaches very large values, either positive or negative, as a result of stimulated scattering to the spin level of lowest energy ($+1/-1$ spin for positive/negative detuning).

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Semiconductor microcavities have attracted increasing interest in the last decade because they allow a precise control of the radiation-matter interaction. This interaction is strongest when the characteristic frequencies of photons (radiation) and excitons (matter) are brought into resonance. Two different regimes can be established under this resonance condition: the strong and weak coupling regimes. The largest effort has been devoted to the study of the strong coupling regime (SCR), in which the eigenstates of the system are no longer pure exciton or photon but a superposition of both, known as cavity polaritons [1,2]. The resonant frequencies of excitons and photons are split, leading to the so-called Rabi splitting, in analogy with atomic cavities [3]. Only in the last few years has it been possible to observe the polariton nonlinear emission in both III-V [4–6] and II-VI [7–10] semiconductor microcavities.

Another issue that has drawn a lot of attention in the nonlinear SCR is the existence of a polariton-polariton scattering mechanism stimulated by the final-state population. This mechanism will be active in a bosonic system, such as cavity polaritons, as soon as the final-state population approaches unity. Clear experimental evidences of this stimulated scattering have been reported recently in the literature [5,11]. In those experiments, the parametric scattering was enhanced by a convenient choice of the angle of incidence of the excitation beams. The result is a macroscopic polariton occupancy ("condensation") of the states at $\mathbf{K} \sim 0$ and $\mathbf{K} \sim 2\mathbf{k}_{\text{pump}}$, where \mathbf{k}_{pump} is the incident pump wave vector.

Concomitantly, a rekindled interest on the carriers' spin in semiconductor structures has given rise to a new field, spintronics, which explores the possibility of designing new spin-based devices, useful for advanced applications such as optical memories and switches, quantum cryptography, and quantum computing. The degree of circular polarization of the emission is directly related to the spin of the elementary excitations of the system, defined as the third component of the total angular momentum. The spin

relaxation processes of excitons, electrons, and holes have been extensively studied in the last decade [12–15]. In the particular case of cavity polaritons, due to the mixed photon-exciton character, significant changes on their spin dynamics with respect to bare quantum wells are expected. Nevertheless, the spin has been considered only very recently [11,16–18].

In this Letter, the fundamental issue of polariton spin dynamics is investigated under nonresonant excitation conditions, which resemble those expected in real devices. We demonstrate that the polariton spin plays a crucial role in the stimulated scattering process, which could lead to an exciton-polariton condensate and polariton lasing. We establish that semiconductor microcavities offer unique possibilities to control the helicity of the light emission, what could be exploited to develop ultrafast optical polarization switches. This control, determined by the exciton-cavity detuning, results from a breaking of the degeneracy of the spin-up and spin-down cavitylike states: the ground state of the system is spin-up (spin-down) at positive (negative) detunings.

The sample under study is a $\lambda/2$ $\text{Cd}_{0.40}\text{Mg}_{0.60}\text{Te}$ microcavity. A slight wedge in the cavity thickness, obtained by suppression of sample rotation during molecular-beam-epitaxy growth, allows tuning of the cavity and the exciton by moving the excitation spot across the wafer. In the antinode position of the electromagnetic standing wave two CdTe QWs of 90\AA are placed. The top/bottom cavity mirrors are distributed Bragg reflectors (DBRs) made of 17.5/23 pairs of alternating layers of $\text{Cd}_{0.40}\text{Mg}_{0.60}\text{Te}$ and $\text{Cd}_{0.75}\text{Mn}_{0.25}\text{Te}$. The cavity finesse, extracted from cw-reflectivity measurements, amounts to ~ 1200 , assuring the excellent quality of the sample. The Rabi splitting characteristic of the microcavity is also determined by cw-reflectivity measurements and amounts to ~ 10.5 meV.

The experiments are made in backscattering geometry with pulses provided by a Ti:sapphire laser, and the sample kept at 5 K. The photoluminescence (PL) is collected, for $K \leq 1 \times 10^4 \text{ cm}^{-1}$, and time resolved using an

up-conversion spectrometer with 2 ps resolution. For polarization-resolved measurements, $\lambda/4$ plates are used to analyze the emitted PL into its σ^+ - and σ^- -polarized components, after excitation with σ^+ light pulses. The circular degree of polarization of the PL is defined as $\varphi = \frac{I^+ - I^-}{I^+ + I^-}$, where $I^{+/-}$ is the intensity of the $\sigma^{+/-}$ component of the PL. φ is denoted in the following as polarization. The nonresonant excitation energy is tuned to the first reflectivity dip above the stop band of the DBRs, ~ 90 meV higher than the emission energy of the cavitylike polariton branch, to assure the same excitation conditions in all the experiments. The created excitons relax very fast (~ 100 fs) by optic phonon emission to polariton states. The scattering processes to thermalize the polaritons close to $\mathbf{K} \sim 0$ are profusely discussed in Refs. [7,19], where exciton-exciton Coulomb scattering is shown to play a key role.

We have studied the time evolution of the polariton PL as a function of the cavity-exciton detuning ($\delta = E_C - E_X$) and of the average excitation power density. Figure 1 shows a contour plot that compiles the PL spectra at 10 ps, measured at different points of the sample. The PL shows a clearly resolved doublet, which allows us to determine the energy splitting between the two normal modes. A clear anticrossing between the bare states is observed, leading to a Rabi splitting of ~ 9.5 meV, comparable to that observed in cw experiments. We concentrate on the results at $\delta = \pm 10$ meV, which show the highest values of φ . At intermediate detunings the dynamics of the light emission evolves monotonously between these two cases; however, φ presents a rich evolution, with striking phenomena such as the existence of a time plateau, which will be presented elsewhere [20].

The integrated intensity of the photonlike branch emission in a point of the sample characterized by $\delta \sim -10$ meV is depicted in the inset of Fig. 1, for a delay time of ~ 20 ps. An exponential rise of the emission intensity with increasing excitation density is observed above 7.5 W/cm². A similar exponential growth has been reported and interpreted in terms of final-state stimulated scattering, as is characteristic of a bosonic system when the final-state ($\mathbf{K} \sim 0$) occupancy approaches unity [21]. Our results provide an experimental evidence of a very efficient polariton-polariton stimulated scattering, even under nonresonant excitation. In the case of positive detuning the excitation power dependence is more complicated. For small excitation densities the emission arises mainly from the LPB states. However, a crossover is observed around 7 W/cm², when the emission of the upper polariton branch (UPB) exceeds that of the LPB. A similar exponential growth of the integrated intensity versus excitation density is then observed, indicating the existence of a stimulation process.

In the following we consider only the nonlinear emission regime and concentrate on the spin dynamics of cavity polaritons for both positive and negative detunings. The

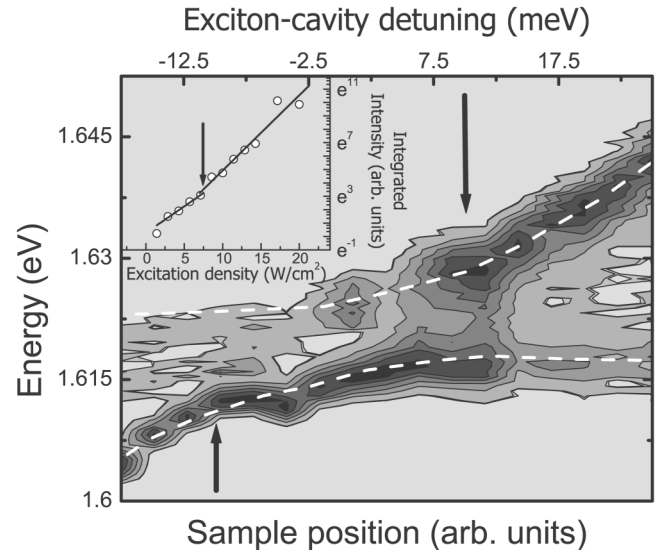


FIG. 1. Contour plot of the PL at 10 ps measured in different points of the sample. Black indicates high intensity; light gray indicates low intensity. The dashed lines are guides to the eye. The arrows indicate the detunings discussed in the text. Inset: integrated intensity (log scale) of the lower polariton branch (LPB) emission at 20 ps as a function of the excitation power density for $\delta = -10$ meV. The arrow points to the threshold for observing nonlinear effects in the emission.

polarization-resolved time evolution of the emission from the cavitylike polariton states for an excitation density of 18 W/cm² is shown in Figs. 2(a) and 2(b) for a detuning of $+10/-10$ meV. For $\delta > 0$ [Fig. 2(a)], the emission intensity of the σ^+ component of the PL (solid circles) is much bigger than that of the σ^- -polarized one (open circles). This intensity difference gives rise to a nonvanishing polarization of the emission, whose time evolution is depicted in Fig. 2(c). The initial polarization is $\sim 30\%$ and it increases up to $\sim 90\%$ at ~ 15 ps. This rise of φ , which has been also reported recently for III-V microcavities [17,18], is markedly different to the monotonous decrease observed for bare excitons. It can be qualitatively understood considering that the relaxation to $\mathbf{K} \sim 0$ states, driven by the polariton-polariton stimulated scattering, is spin selective and occurs in a much shorter time scale than any spin relaxation. This will imply that the slightly unbalanced $+1$ spin population, created at $\mathbf{K} > 0$ by a σ^+ -polarized pulse, is mirrored at $\mathbf{K} \sim 0$, creating a seed for stimulation only of the $+1$ spin population. However, this argument alone is not sufficient to explain the results for $\delta < 0$ as we show below. After reaching its maximum value, φ decreases to zero, as a result of two different processes. The first one is the rapid σ^+ -polarized stimulated emission, which results in a considerable reduction of the $+1$ spin population. The second process, slower than the first one, is the conventional spin relaxation, which tends to equalize both spin populations.

The behavior is quite different for $\delta < 0$. The polarization-resolved time evolution of the cavitylike

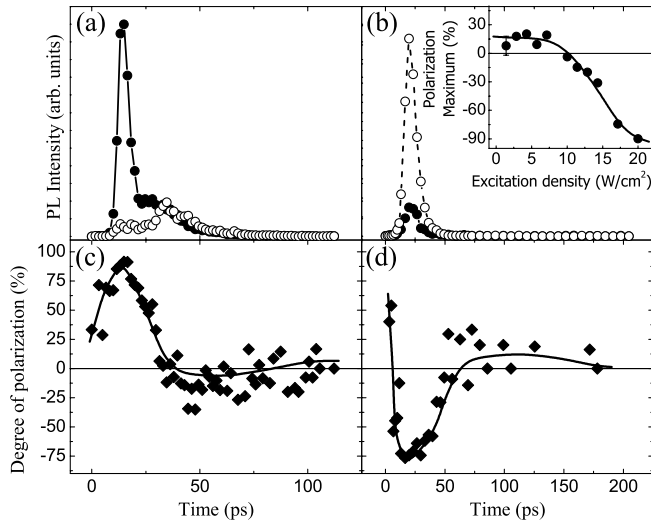


FIG. 2. (a) Time evolution of the circularly polarized PL of the UPB at $\delta = 10$ meV. The filled circles/solid line (open circles/dashed line) denote the σ^+ (σ^-) emission. (b) Same as in (a) for the LPB at $\delta = -10$ meV. Inset: maximum value of the polarization degree at 20 ps as a function of excitation density for $\delta = -10$ meV. The line is a guide to the eye. (c) Time evolution of the circular polarization degree of the PL emission of the UPB at $\delta = 10$ meV. (d) Same as in (c) for the LPB at $\delta = -10$ meV. The data are taken with an excitation density of 18 W/cm^2

mode emission [Fig. 2(b)] shows that the intensity of the σ^- emission (open circles) is larger than that of the σ^+ -polarized one (solid circles). Therefore, the emission is counterpolarized with the excitation. This is made evident in the time evolution of φ , depicted in Fig. 2(d). The initial value of φ is $\sim 50\%$, but it changes very rapidly to $\sim -75\%$ at ~ 20 ps. With increasing excitation density φ reaches very large negative values, saturating at $\sim -90\%$ for excitation densities larger than 20 W/cm^2 [see the inset of Fig. 2(b)]. The σ^+ -polarized excitation still creates a larger +1 spin population, which is reflected as a positive φ at $t = 0$. However, the scattering to $\mathbf{K} \sim 0$ states of -1 spin becomes more efficient at longer times ($t \sim 20$ ps), resulting in a larger -1 spin population at $\mathbf{K} \sim 0$ and therefore to a counterpolarized (σ^-) emission.

In order to explain the negative values of polarization, one could argue that changing the excitation energy, going from $\delta > 0$ to $\delta < 0$, a resonant excitation condition with the light-hole excitons is met. However, this explanation can be disregarded because the excitation energies in our experiments are always above the light-hole exciton (at least 30 meV) and, furthermore, a negative value of φ at $t = 0$ would be obtained. The qualitative argument used to understand the spin dynamics for $\delta > 0$ does not apply anymore for the case of $\delta < 0$, since the seed for stimulation would still have +1 spin, resulting in positive values of φ .

A detailed study of the polarization-resolved PL spectra clarifies the origin of the different spin dynamics for posi-

tive and negative detunings: a small energy splitting ($\Delta = E^- - E^+$, where $E^{+/-}$ denotes the $\sigma^{+/-}$ emission energy) between the σ^+ and the σ^- components of the PL is obtained at short delay times, as shown in Fig. 3. This splitting evidences that the +1 and -1 spin states are no longer degenerate in energy at $\mathbf{K} = 0$.

The polarization-resolved PL spectra at 20 ps delay for $\delta = 10$ meV are depicted in Fig. 3(a): the σ^+ emission (filled circles, solid line) occurs at lower energy than the σ^- -polarized one (open circles, dashed line). The spin splitting, Δ [inset of Fig. 3(a)] increases with excitation density, saturating at ~ 1 meV. On the contrary, in the case of $\delta < 0$ (Fig. 3 b), the σ^- component lies at lower energies, revealing that the -1 spin state is the lowest energy state at very short times. In this case Δ is negative and saturates at ~ 0.5 meV [inset of Fig. 3(b)].

Let us describe in more detail the relaxation process of the nonresonantly created excitons towards $\mathbf{K} = 0$ states, taking into account the $+1/-1$ spin splitting, and the fact that polariton pair scattering is spin selective [11,16–18].

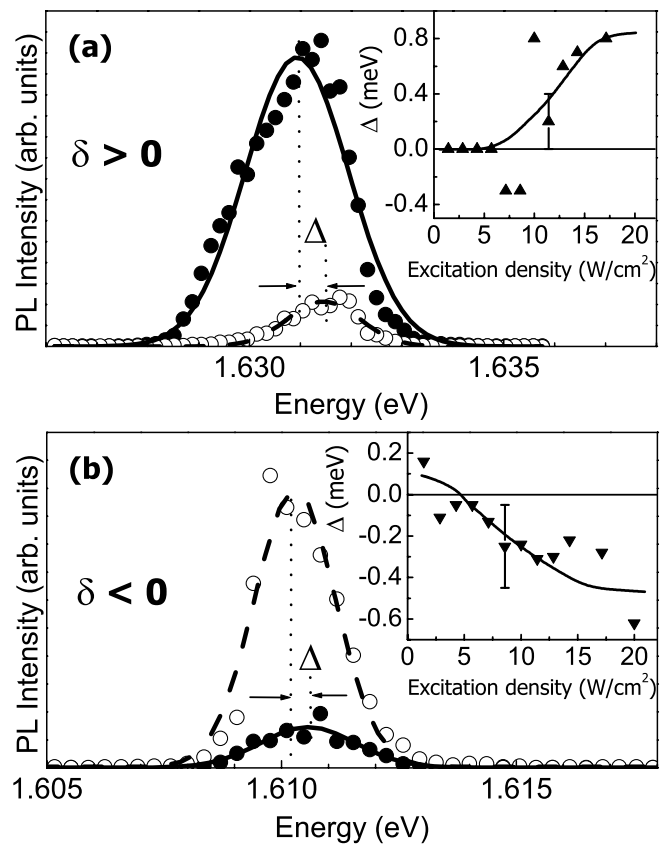


FIG. 3. (a) Polarization-resolved PL spectra at 20 ps for an excitation density of 18 W/cm^2 at $\delta = 10$ meV. The filled circles/solid line (open circles/dashed line) denote the σ^+ (σ^-) emission. The lines are Gaussian fits of the experimental data. Inset: Spin splitting (Δ) at 20 ps as a function of excitation density for $\delta = 10$ meV. The line is a guide to the eye. (b) Same as in (a) for $\delta = -10$ meV.

For $\delta > 0$, the large wave vector excitonlike polaritons from the LPB states are scattered to the cavitylike UPB $\mathbf{K} = 0$ states. Most of the polaritons are transferred to the lowest energy spin level, i.e., $+1$ states, creating the seed for stimulation. This accumulation of $+1$ spin polaritons results in a large σ^+ -polarized stimulated emission and a considerably smaller σ^- -polarized one: \wp is positive and becomes very large at short times [see Fig. 2(c)]. The $+1$ spin state is emptied very quickly through the σ^+ -polarized stimulated emission and therefore the polarization decreases to zero very rapidly after reaching the maximum. This balance of the populations, which is reinforced by the conventional spin relaxation processes, equalizes the intensities of both circularly polarized components of the PL and \wp remains at zero.

For $\delta < 0$, the lowest energy level at $\mathbf{K} \sim 0$ is now the -1 spin state. The accumulation of polaritons in those states results in a large σ^- emission. Therefore \wp becomes negative and very large at short times. Similarly to the $\delta > 0$ case, now the -1 spin population is rapidly reduced and \wp goes back to zero.

The physical origin of this energy splitting between the two spin states at $\mathbf{K} \sim 0$ still needs to be clarified, but it is likely to account for the reversal of the circular degree of polarization of the PL with changing the exciton-cavity detuning. The splitting would be compatible with a decrease in the light-matter interaction strength for the majority polaritons ($+1$), which are initially created by the σ^+ -polarized excitation, as compared to that of the minority (-1) polaritons. This would imply that for negative (positive) detuning, the $+1$ states would lie above (below) the -1 , rendering a $\Delta < 0$ (> 0) as borne out by our results. However, our experiments show that with increasing excitation density, an initial blueshift of 0.5 meV for both ± 1 states, without any splitting, is followed by a redshift of the -1 polaritons, while the $+1$ remain at the same energy. Therefore, the coupling strength of the $+1$ polaritons does not decrease, invalidating the previous argument. The fact that the splitting increases with excitation power density indicates that it could originate from exciton-exciton interactions. An existing 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 [22], but only for $\Delta < 0$. Further experiments are under way to understand this spin splitting and its \mathbf{K} dependence.

In summary, we have investigated the relaxation and the spin dynamics of cavity polaritons after nonresonant pulsed excitation, in the nonlinear emission regime. An increase of the excitation density leads to an exponential growth of the integrated emission intensity from the cavity-

like states, providing an experimental evidence for the existence of final-state stimulated scattering. The spin dynamics presents novel phenomena, such as the existence of a maximum at a finite time and a sign reversal of the circular degree of polarization. This reversal is related with the sign of the splitting between the energies of the σ^+ - and σ^- -polarized components of the PL. The spin of the lowest photonlike energy state changes from $+1$ for $\Delta > 0$ to -1 for $\Delta < 0$.

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