PERGAMON

Solid State Communications 117 (2001) 267–271

solid
state
communications

www.elsevier.com/locate/ssc

Spin dynamics of cavity polaritons

M.D. Martín[a], L. Viña[a,*], J.K. Son[b], E.E. Mendez[b]

[a]Departamento de Fisica de Materiales C-IV, Universidad Autonoma de Madrid, Cantoblanco, E-28049 Madrid, Spain
[b]Department of Physics and Astronomy, SUNY at Stony Brook, NY 11794-3800, USA

Received 6 September 2000; accepted 25 September 2000 by F. Yndurâin

Abstract

We have studied polariton spin dynamics in a GaAs/AlGaAs microcavity by means of polarization- and time-resolved photoluminescence spectroscopy as a function of excitation density and normal mode splitting. The experiments reveal a novel behavior of the degree of polarization of the emission, namely the existence of a finite delay to reach its maximum value. We have also found that the stimulated emission of the lower polariton branch has a strong influence on spin dynamics: in an interval of ~150 ps the polarization changes from +100% to negative values as high as −60%. This strong modulation of the polarization and its high speed may open new possibilities for spin-based devices. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: A. Quantum wells; D. Optical properties; D. Spin dynamics; E. Time-resolved optical spectroscopies

PACS: 71.36. + c; 78.45. + h; 78.47. + p; 71.35.Gg

Semiconductor microcavities are one of the most suitable structures to study lightmatter interaction. In the strong coupling regime, first observed in 1992, excitons and photons form mixed states, named cavity polaritons [1]. The signature of this regime is an anticrossing of the exciton and cavity modes when they are brought into resonance. Although cavity polaritons have been profusely investigated [2–5], there are some aspects of their optical properties which need better understanding, in particular those that refer to non-linear processes and the polarization of the emitted light.

In studying non-linear processes, it has been difficult to maintain the polaritonic signature because of saturation of the strong coupling regime [5–8]. This is the case in Vertical Cavity Surface Emitting Lasers (VCSEL's), where the non-linear emission originates in the population inversion of a dense electron–hole plasma in the weak coupling regime [9]. The polaritonic character of stimulated emission in both III–V [10] and II–VI [11,12] microcavities reported recently has been questioned by calculations that claim that those results can be explained within a fermionic quantum theory [8].

The polarization of the light emitted by bare semiconductor quantum wells (QWs) has been widely investigated [13,14]. In fact, because the polarization is directly linked to the spin (i.e. the third component of the total angular momentum), its study is part of a new field, known as spintronics that aims to develop spin-based fast optoelectronic devices. The mechanisms responsible for the spin relaxation of excitons in QWs and its dependence on different parameters such as well width, temperature and excitation density have been established [13–15]. In microcavities, due to the mixed photon–exciton character of the polaritons and the inefficiency of those mechanisms on the cavity like mode, significant changes on the spin dynamics are to be expected. However, in spite of these differences, only a few works on the polarization properties of VCSEL's [16,17] and microcavities [18,19] have been reported. Very recently, the polarization properties of semiconductor microcavities have been studies both, in cw-PL [18] and in pump and probe reflectivity experiments [19]. Our work complements the novel results by Savvidis et al. [19] of coherent gain in microcavities, and its spin dependence, which takes place in a very short time scale, showing that these effects are also present at considerably longer times, in the incoherent regime. Following a preliminary study of spin dynamics in a semiconductor microcavity [20], we report in this paper on the time evolution of the polariton

---

[*] Corresponding author. Tel.: +34-91-397-4782; fax: +34-91-397-8579.
E-mail address: luis.vina@uam.es (L. Viña).

PII: S0038-1098(00)00426-9

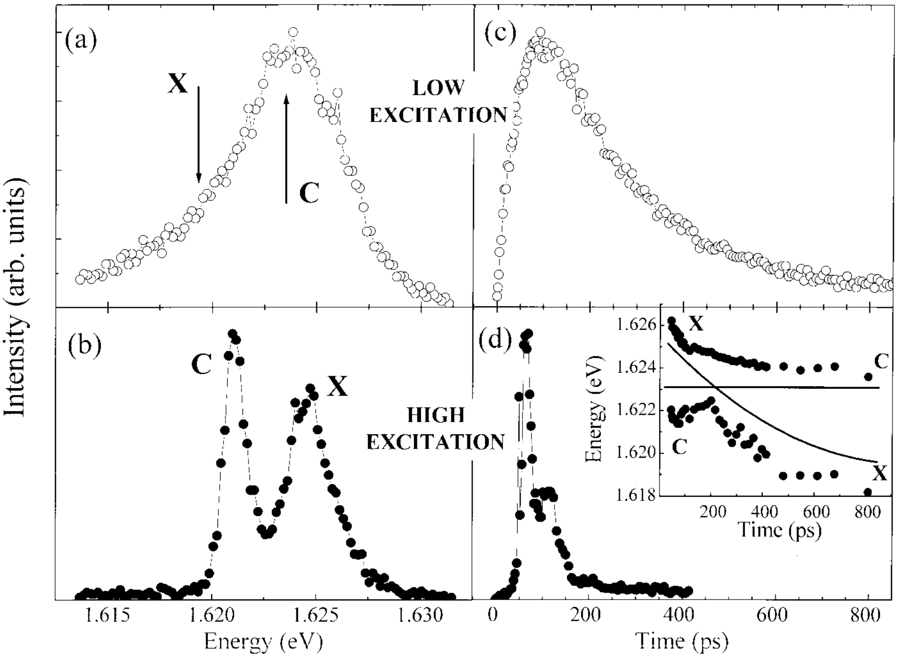

Fig. 1. (a) PL spectra measured 100 ps after excitation with a $0.35I_{th}$ density. (b) PL spectra measured 100 ps after excitation with an I_{th} density. (c) Cavity-mode evolution for an excitation density of $0.35I_{th}$. (d) Cavity-mode time evolution for an excitation density of I_{th}. Inset: energy positions of both polariton branches as a function of time for a density I_{th}. The lines are a guide to the eye.

spin in both linear and non-linear regimes. Specifically, we show that the emission is highly polarized in the non-linear regime and that the polarization dynamics is strongly influenced by exciton-cavity detuning.

The microcavity studied in this work, grown by molecular beam epitaxy, consists of three GaAs quantum well regions embedded in a $3\lambda/2$ $Al_{0.25}Ga_{0.75}As$ Fabry–Perot resonator clad by dielectric mirrors. The top and bottom mirrors are distributed Bragg reflectors made of twenty and a half and twenty-four alternating $AlAs/Al_{0.35}Ga_{0.65}As$ $\lambda/4$ layers, respectively. The QWs are placed at the antinodes of the resonator's standing wave. A slight variation (introduced by design during growth) of the cavity's thickness along the radial direction of the wafer allowed to tune the cavity's resonance with the transition in the QWs by moving the excitation spot across the sample. Using low temperature cw-photoluminescence (PL) measurements we have identified exciton-like (X) and cavity-like (C) modes, whose Normal Mode Splitting (NMS) variation was found to be between 3.5 and 7 meV as the laser spot was moved across the sample. The small width (4 meV) of the X-like polariton PL peak and its high intensity attest to the high quality of our heterostructures [21].

We have used time-resolved spectroscopy to study polariton recombination and spin dynamics as a function of excitation density and exciton-cavity detuning ($E_C - E_X$). The experiments were performed under non-resonant excitation above the cavity stop-band (1.71 eV) and the PL emitted by the sample was analyzed in a conventional up-conversion spectrometer [22] with a time resolution of ~2 ps. The sample was mounted on a cold-finger cryostat where the temperature was kept at 5 K. For polarization resolved measurements two $\lambda/4$ plates were included in the optical path of the experiment. Under σ^+ excitation, the degree of polarization of the PL is defined as $\wp = (I^+ - I^-)/(I^+ + I^-)$, where $I^\pm$ denotes the PL emitted with ±1 helicity. The analysis of this quantity gives direct information about the spin relaxation processes, as it is directly related to the difference of +1 and −1 spin populations [15]. In the following, we will refer to this quantity simply as polarization.

Initial time-resolved experiments under weak excitation ($I_{exc} < 19$ W/cm^2) showed that the NMS was resolvable for time delays larger than ~300 ps and confirmed the NMS variation across the sample extracted from cw measurements [21]. The study of the time evolution of both peaks revealed that the NMS does not influence the dynamics for positive detunings, in agreement with Abram et al. [23]. The characteristic rise and decay times of the PL were very similar for both polariton branches, and amounted to $\tau_r^X \sim 100$ ps, $\tau_d^X \sim 300$ ps, $\tau_r^C \sim 70$ ps, and $\tau_d^C \sim 250$ ps, where r (d) denotes rise (decay) time.

With increasing excitation density, drastic changes were observed in the time-resolved spectra as well as in the recombination dynamics. At low power densities, both the lower (LPB) and upper (UPB) polariton branches have a

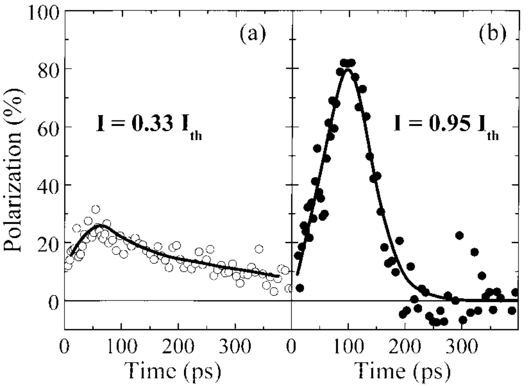

Fig. 2. Time evolution of the cavity-mode polarization for excitation densities: (a) $0.33I_{th}$; and (b) $0.95I_{th}$.

similar dependence on power (slightly larger that linear). This dependence is maintained for the UPB in the whole range of excitation densities used in our experiments (up to 40 W/cm^{-2}, which corresponds to an injected photon density of $\sim 5\times10^{11}$ photons pulse^{-1} and cm^{-2}). In contrast, the dependence of the LPB emission on power shows a threshold, I_{th}, at ~20 W/cm^2 [24].

Fig. 1a and b display two PL spectra measured 100 ps after excitation, below (○, 7 W/cm^2) and at the threshold (●, 20 W/cm^2), respectively. At low power, the C-like mode is at higher energy that the X-like, i.e. $\delta > 0$, similar to the situation found in cw experiments. Although the LPB is not clearly resolved at 100 ps, the NMS becomes apparent at longer times, when both branches have comparable intensities. At high powers, the situation is reversed and the LPB/UPB has photonic/excitonic character, i.e. $\delta < 0$. The LPB can be clearly resolved for excitation densities $I \geq 14$ W/cm^2 as a very narrow peak at 1.621 eV (marked as C in Fig. 1b), and it gets sharper with increasing density, up to I_{th}, reducing its width by a factor of ~4. This narrowing also occurs for the UPB, but its linewidth is only reduced by a factor for ~2. The NMS is practically independent of power.

The time evolution is also strongly affected by an increase of excitation density, as shown in Fig. 1c and d. For small excitation densities (○, 7 Q/cm^2) the time evolution is similar to that typical of QWs under non-resonant excitation: the emission is characterized by slow rise and decay times [13,14]. For larger excitation (●, 20 W/cm^2), the rise and decay times are faster and a delay time of ~30 ps is needed to reach the non-linear emission regime. This onset in the PL might be related with the bottleneck in the relaxation of polaritons towards $\mathbf{K}_{\|} = 0$ states [25]. The rapid rise of the PL observed beyond the threshold could be interpreted in terms of stimulated scattering, which enhances the build up of the polariton $\mathbf{K}_{\|} = 0$ population.

The subject of non-linear emission in semiconductor microcavities has been controversial regarding the existence of polaritons, due to bleaching at high densities [6–8, 10,18,19]. In our experiments, excitons and photons seem to be still strongly coupled above I_{th}, as can be inferred from the existence of an anticrossing (inset of Fig. 1d). The non-resonantly created excitons relax their energy very rapidly towards $\mathbf{K}_{\|} \simeq 0$ states and, a few picoseconds after excitation, the X mode is observed at 1.626 eV. At these very short times, the LPB (1.622 eV) is photon-like. With increasing delay, the X mode red shifts due to the decrease of polariton density, similarly to the behavior of excitons in bare QWs [26]. However, since the C mode energy is density independent, both modes become resonant, and a clear anticrossing is observed at ~180 ps. At longer times, the LPB (UPB) recovers the X-like (C-like) character determined by cw measurements.

The three effects discussed above, linewidth reduction, excitation density threshold and, especially, the anticrossing in time suggest that the PL observed above I_{th} can be attributed to polariton stimulated emission.

Let us now concentrate on polariton spin dynamics. A σ^+ excitation pulse will initially populate the +1 spin level but a −1 spin population will appear as a result of spin flip mechanisms, which eventually balance both spin populations [15] and therefore reduce the polarization to $\wp = 0$. For excitons in bare QWs, the polarization reaches its maximum value just after excitation and then decays exponentially to zero [13,14]. On the other hand, in microcavities, due to the complex nature of polaritons, one expects the spin dynamics of this mixed state to be different from that of bare excitons or photons. This fact is documented in Fig. 2, which depicts the time evolution of the polarization of the cavity mode for two different excitation densities below the nonlinear emission threshold. In contrast with the monotonically decreasing behavior of $\wp$ found in bare QWs, in our microcavity a maximum is observed at a finite time after excitation. The polarization at $t = 3$ ps is ~10%, which means that, after the relaxation of polaritons to $\mathbf{K}_{\|} \simeq 0$ states, only 55% of the total population is in the +1 spin state. Such a small value of $\wp$ is mainly due to the non-resonant excitation conditions. $\wp_{max}$ is reached in 60–100 ps, and its value increases with excitation density, being as high as 80% (Fig. 2b, 19 W/cm^2) as the stimulated emission regime is reached. The direct relation between the time scales observed in the emission and the polarization, under high excitation, reveals the strong influence of the stimulation processes on spin dynamics. These findings also corroborate recent results that show that the polariton system can be markedly spin polarized [18,19].

The fact that a finite is needed to reach $\wp_{max}$ implies that there must be a new scattering mechanism that favors polaritons with +1 spin, and thus competes with spin relaxation and tends to prevent equalization of both spin populations. At very low powers, the relaxation of large in-plane wave vector excitons is governed by the emission of acoustic phonons, which has no spin dependence. At high powers, the new mechanism can be interpreted as polariton–polariton scattering, which is stimulated by the polariton

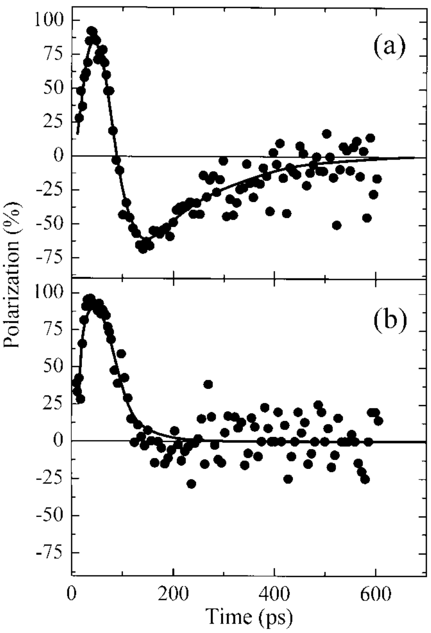

Fig. 3. Time evolution of the cavity-mode polarization for excitation densities $\sim 2I_{\mathrm{th}}$ measured at different points of the sample, characterized by a NMS of: (a) 4.5 meV; and (b) 6 meV.

final-state population [27]. The increase of $\wp_{\max}$ with excitation density (25% at $0.33I_{\mathrm{th}}$, 80% at $0.95I_{\mathrm{th}}$) evidences that there is an enhancement of the scattering to the +1 spin state. The stimulation does not occur for the σ^- polarized LPB emission, which also shows a time evolution with rise and decay times much longer than those observed in the non-linear regime for the σ^+ emission. This process is not only spin selective but also induces an increase of the +1 population by flipping the spin of minoritary polaritons (−1).

An additional fact evidences the importance of the "polaritonic" stimulation on the spin behavior: the decrease in the time needed to reach $\wp_{\max}$ with increasing positive detuning. This means that as we move away from resonance and the excitonic component of the LPB increases, the time evolution of $\wp$ approaches that characteristic of bare excitons, with the maximum value of $\wp$ occurring closer to $t = 0$. The explanation given here is only qualitative and a complete theoretical description must be developed before this new mechanism of scattering into spin polarized states can be fully understood.

Recently, a rate-equation model has been successfully applied to describe the optical properties of cavity-polaritons in the nonlinear regime, under cw excitation [6,7], and a microscopic fermionic many-body theory has been developed to explain the linear and non-linear behavior of normal-mode coupling in microcavities, including dynamics of the light emission [8]. However, even in these models, the spin of the polaritons was not taken into account. Our results provide new valuable information on the stimulated scattering into spin-polarized states of the LPB, and should attract interest for theoretical work that includes the spin in the calculations.

For excitation densities above the threshold, the time evolution of the cavity-mode polarization displays a behavior even more surprising. The LPB polarization reaches values as high as 95% when entering into the nonlinear emission regime. In contrast, for the UPB, although it shows a similar behavior, its polarization is only 60%. After the initial rise of the polarization, once the maximum is reached, its dynamics is strongly dependent on NMS. Fig. 3 depicts the time evolution of $\wp$ for two different points of the sample, with different NMS, under an excitation density of 2 I_{th}. For small exciton-cavity detunings (Fig. 3a, 4.5 meV) a negative dip (−60%) is observed at ~150 ps, which is absent for larger NMS (Fig. 3b, 6 meV).

The negative polarization is a consequence of the fast disappearance of the +1 polaritons, due to the stimulated σ^+ emission of the LPB and the concurrent slower dynamics of the σ^- emission. The −1 polariton population overcomes that of +1 spin due to the lack of stimulation for σ^- polarization and also because the spin–flip processes are not fast enough to compensate the emptying of the +1 polaritons. The remarkable change in the state of polarization of the emitted light from +80% to −60%, taking place in a very short time (~100 ps), is unique and, to the best of our knowledge, has not been reported before in any semiconductor based system.

Once the minimum of $\wp$ is reached, the polarization dynamics becomes slower: by then the polariton population has decreased by a factor 5–10 (depending on power density) and the remaining +1 spin population is too small to give rise to stimulation. Under these conditions, only the usual spin–flip mechanisms govern the polarization, which decreases steadily. Fig. 3b shows that the negative dip has disappeared for larger NMS, due to the modification of the stimulated emission dynamics. The decay time of the σ^+ PL becomes slower and the loss of +1 polaritons is neutralized by flipping −1 spins and as a result $\wp$ does not reach negative values. One can also observe in Fig. 3b that the abrupt decay of the polarization is slowed down with increasing NMS.

It should be mentioned that excitation with σ^- yields identical results to those of the σ^+ excitation discussed above, as expected from time reversal symmetry arguments. The sign reversal of the polarization is also observed for the σ^- excitation and it is also the majority spin population (−1 in this case) the only one that undergoes stimulation.

In summary, our experiments on polariton recombination as a function of excitation density and exciton-cavity detuning have revealed strong nonlinearities in the emission of the lower polariton branch. A careful study of the time evolution of the polarization has shown the existence of a new scattering mechanism for the polaritons that is spin selective and gives rise to very high values of the polarization. The Normal Mode Splitting plays a key role on the spin relaxation of cavity polaritons, leading to a reversal in the polarization for small detunings. The large contrast in the polarization and its high speed open the possibility of new concepts for spintronic devices, such as ultrafast switches, based on the spin dynamics of microcavity polaritons.

Acknowledgements

We are thankful to Dr I.W. Tao and R. Ruf for growing the samples used in this work, which has been supported by Fundación Ramón Areces, the Spanish DGICYT under contract PB96-0085, the CAM (07N/0026/1998), the Spin-US Joint Commission and the US Army Research Office.

References

[1] C. Weisbuch, M. Nishioka, A. Ishikawa, Y. Arakawa, Phys. Rev. Lett. 69 (1992) 3314.
[2] R. Houdré, et al., Phys. Rev. Lett. 73 (1994) 2043.
[3] T.B. Norris, J.-K. Rhee, C.-Y. Sung, Phys. Rev. B 50 (1993) 14663.
[4] B. Sermage, et al., Phys. Rev. B 53 (1996) 16515.
[5] R. Houdré, et al., Phys. Rev. B 52 (1995) 7810.
[6] S. Pau, et al., Phys. Rev. A 54 (1996) R1789.
[7] M. Kira, et al., Phys. Rev. Lett. 79 (1997) 5170.
[8] G. Khitrova, et al., Rev. Mod. Phys. 71 (1999) 1591.
[9] T.E. Sale, Vertical Cavity Surface Emitting Lasers, Wiley, New York, 1995.
[10] P. Senellart, J. Bloch, Phys. Rev. Lett. 82 (1999) 1233.
[11] L.S. Dang, et al., Phys. Rev. Lett. 81 (1998) 3920.
[12] J. Bleuse, et al., J. Cryst. Growth 104–105 (1998) 750.
[13] T.C. Damen, L. Viña, J.E. Cunningham, J. Shah, Phys. Rev. Lett. 67 (1991) 3432.
[14] L. Muñoz, E. Pérez, L. Viña, K. Ploog, Phys. Rev. B 51 (1995) 4247.
[15] M.Z. Maialle, E.A. de Andrada e Silva, L.J. Sham, Phys. Rev. B 47 (1993) 15776.
[16] H. Ando, T. Sogawa, H. Gotoh, Appl. Phys. Lett. 73 (1998) 566.
[17] A. Gahl, S. Balle, M. San Miguel, IEEE J. Quantum Elect. 35 (1999) 342.
[18] A.I. Tartakowskii, et al., Phys. Rev. B 60 (1999) R11293.
[19] P.G. Savvidis, et al., Phys. Rev. Lett. 84 (2000) 1547.
[20] M.D. Martin et al., Proceedings of the 24th International Conference on the Physics of Semiconductivity, 1999.
[21] I.W. Tao, et al., Physica E 2 (1998) 685.
[22] J. Shah, IEEE J. Quantum Elect. 24 (1988) 276.
[23] I. Abram, et al., in: J. Rarity, C. Weisbuch, et al. (Eds.), Microcavities and Photonic Bandgaps: Physics and Applications, NATO Advanced Studied Institute, Series E, Vol. 324, Kluwer Academic, Dordrecht, 1996.
[24] M.D. Martin, et al., Physica E 6 (2000) 169.
[25] F. Tassone, et al., Phys. Rev. B 56 (1997) 7554.
[26] S. Schmitt-Rink, D.S. Chemla, D.B. Miller, Phys. Rev. B 32 (1985) 6601.
[27] T. Baars, et al., Phys. Rev. B 61 (2000) R2409.