phys. stat. sol. (a) **178**, 539 (1999)

Subject classification: 71.35.Gg; 71.36.+c; 78.45.+h; 78.47.+p; S7.12

Spin Polarization Dynamics in a Semiconductor Microcavity

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(Received August 30, 1999)

We have studied the spin dynamics of the polaritons 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, performed under non-resonant excitation, revealed a novel behaviour of the polarization degree of the emission, which reaches its maximum at a finite time. We have also found that polariton stimulated emission takes place for excitation densities larger than 20 W/cm^2. This process has a strong influence on spin dynamics: the emission becomes almost completely polarized and negative values of the polarization are observed, whose magnitude depends on exciton–cavity detuning.

Since the first experimental evidence of microcavity polaritons [1], the electronic properties of semiconductor microcavities have been intensively studied: polariton dispersion [2], relaxation [3] and bleaching [4] have been investigated. In the last years, nonlinear processes have attracted much attention. However, it has proven difficult to keep polaritonic signatures due to strong exciton–cavity coupling saturation [5] (conventional Vertical Cavity Surface Emitting Lasers (VCSELs) work in the weak coupling regime). Only very recently, polaritonic stimulated emission has been reported both in III–V [6] and II–VI [7] microcavities. In the majority of these studies the polariton spin (i.e. third component of the total angular momentum) was not taken into account. There are only a few reports on the spin properties of VCSELs [8]. Our work provides another experimental evidence of polaritonic stimulated emission and evidences the important role played by microcavities on spin properties.

The samples are grown by molecular beam epitaxy and include $Al_{0.35}Ga_{0.65}As$/AlAs dielectric mirrors. Three pairs of GaAs coupled quantum wells (QWs) are placed in the antinode positions of the $3\lambda/2$ $Al_{0.25}Ga_{0.75}As$ microcavity. A slight wedge in the cavity thickness allows tuning the cavity resonance to the transition in the QWs by moving the excitation spot across the sample. Temperature dependent cw-photoluminescence (PL) experiments allow us to attribute an excitonic character to the low-energy-side peak, and a photonic character to the high-energy-side one. Additional magneto-PL experiments confirmed this assignment [9].

Our experiments are performed under non-resonant excitation, above the cavity stop band, and the PL emitted by the sample is analysed in a conventional up-conversion

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spectrometer with a time resolution of ≈2 ps. For polarization resolved measurements a pair of $\lambda/4$ plates are included in the experiment. The samples are mounted in a cold finger cryostat and a temperature of 5 K is kept constant through all the experiments. We have studied the time evolution of both the PL and its polarization degree, as a function of excitation density and exciton–cavity detuning.

For low excitation densities, we have found normal mode splitting variation between 3.5 and 7 meV probing different points on the sample. Our experiments reveal no influence of the exciton–cavity detuning on the PL characteristic rise and decay times, in agreement with [10]. These characteristic times are similar for both polariton branches and amount to $\tau_r^X \approx 100$ ps, $\tau_d^X \approx 300$ ps, $\tau_r^\gamma \approx 70$ ps and $\tau_d^\gamma \approx 250$ ps, where X (γ) and r (d) denote exciton- (photon-)like and rise (decay) time, respectively.

Increasing excitation density leads to a strong modification of the light emission dynamics. Figure 1 depicts two time-resolved PL spectra taken 100 ps after excitation, for small (filled circles, 9 W/cm^2) and large (open circles, 25 W/cm^2) excitation densities. With increasing power, the linewidth of both polaritons is strongly reduced, specially that of the lower polariton branch (LPB), which changes from 7 to 1.5 meV (the change in the upper polariton branch (UPB) amounts only to 2 meV). The inset shows the integrated emission of the LPB (on logarithmic scale) as a function of excitation density: a large increase is seen for excitation densities larger than 20 W/cm^2. This threshold density increases with increasing normal mode splitting.

The time evolution is also drastically modified by excitation density, as demonstrated in Fig. 2. Under small excitation densities (filled circles, 9 W/cm^2) the time evolution is similar to that typical of QWs under non-resonant excitation: the emission begins at zero delay and is characterised by slow rise- and decay-times. For larger excitation densities (open circles, 25 W/cm^2) an onset of the PL at ≈30 ps is observed, which is followed by a very fast rise and decay. This finite time to obtain a noticeable PL is related to the accumulation and relaxation of non-resonantly created excitons towards energies where they can interact with the cavity. The curvature of the initial rise of the PL time evolution shows a threshold similar to that of the LPB integrated emission.

A clear anticrossing between both polariton branches is observed as a function of time (inset of Fig. 2). At short times the UPB is exciton-like

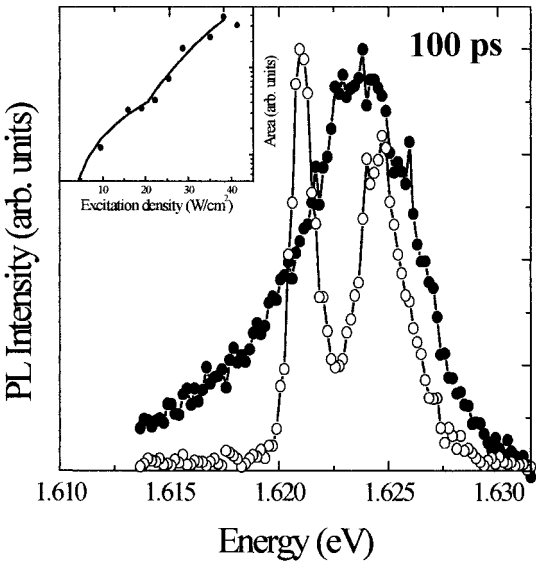

Fig. 1. Time-resolved PL spectra taken 100 ps after excitation, for excitation densities below (9 W/cm^2, ●) and above (25 W/cm^2, ○) the threshold for stimulated emission. Inset: Integrated emission (in logarithmic scale) of the LPB at 100 ps versus excitation density

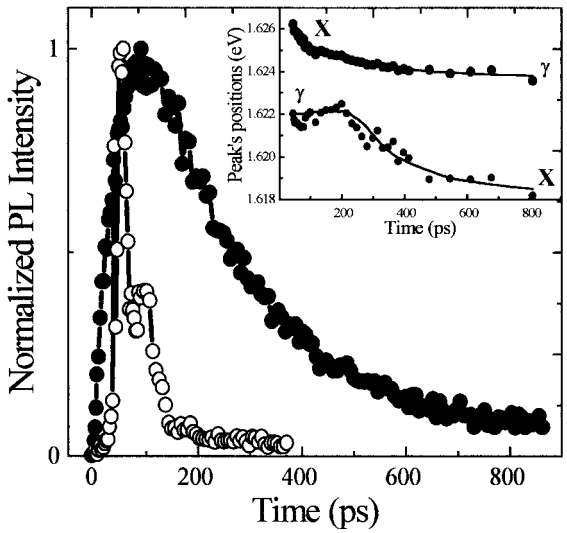

Fig. 2. Time evolution of the cavity mode for excitation densities below (9 W/cm^2, ●) and above (25 W/cm^2, ○) the threshold for stimulated emission. Inset: Energies of both polariton branches as a function of time. The lines are guides to the eye

while the LPB is photon-like. The energy of the exciton-like polariton-mode decreases with time, while that of the cavity-like remains constant. At long times, the LPB (UPB) recovers its exciton- (photon-) like character. This, in addition to the linewidth reduction, the time evolution modification and the excitation density threshold, lead us to identify the non-linear luminescence as polaritonic stimulated emission.

Let us now focus on the polariton spin dynamics. Under σ^+ excitation, the PL polarization degree $\wp$ is defined as

$$\wp = \frac{I^{\sigma^+} - I^{\sigma^-}}{I^{\sigma^+} + I^{\sigma^-}} \tag{1}$$

where $I^{\sigma^{+/-}}$ denotes the PL emitted with ± 1 helicity. An analysis of this magnitude gives direct information about the spin relaxation, as it is directly related to the difference between +1 and −1 spin populations. A σ^+ excitation pulse will *mainly* create +1 excitons and a −1 population will appear as a result of spin-flip processes [11].

Our experiments reveal that the time evolution of $\wp$ in a microcavity is quite different to that characteristic of isolated QWs. For bare excitons, $\wp$ reaches its maximum almost immediately after excitation and then decays exponentially to zero [12]. Under far, non-resonant, excitation $\wp$ should be small, because both kinds of excitons, ± 1, will be created. However, we observe a maximum in $\wp$ a few tenths of picoseconds after excitation. Figure 3 shows several time evolutions of the polarization degree of the cavity mode for two different exciton–cavity detunings and for excitation densities below (filled circles) and above (open circles) the stimulated emission threshold. Let us first concentrate on the behaviour of $\wp$ at moderate excitation densities (Fig. 3, filled circles). The initial value of $\wp$ is only $\approx$10% and then rises up to $\approx$40% at 50 to 100 ps after excitation. The time delay to reach the maximum increases with decreasing exciton–cavity detuning. This maximum value increases strongly with excitation density, reaching values as high as 70% before entering the stimulated emission regime. The fact that a finite time is needed to reach $\wp_{\max}$ implies that the interaction with the cavity field favours +1 polaritons at the expense of the −1 population. This process competes with spin relaxation, which tends to make both populations equal.

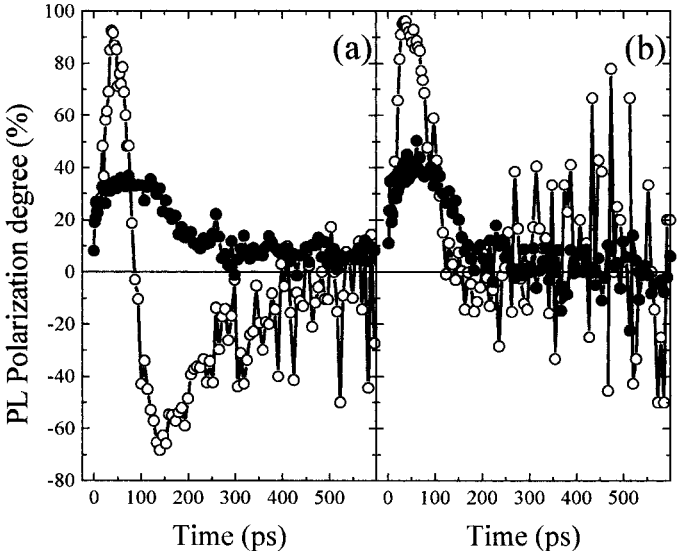

Fig. 3. a) Time evolution of the polarization degree of the cavity mode for 16 (●) and 40 W/cm^2 (○), in a point of the sample with a normal mode splitting of 4.5 meV. b) Time evolution of the polarization degree of the cavity mode for 25 (●) and 45 W/cm^2 (○), for a normal mode splitting of 6 meV

For excitation densities well above the threshold (Fig. 3, open circles) the dynamics of $\wp$ is even more conspicuous. The initial value of $\wp$ is ≈30% rising up to 95% at ≈30 ps. This means that the initial +1 polariton population increases from 65% to ≈97%. The UPB displays a similar behaviour but the effect is much smaller ($\wp$ maximum is ≈60%). Although detuning does not affect the value of $\wp_{max}$, it drastically modifies the temporal evolution of $\wp$ after the maximum is reached. For small normal mode splitting (Fig. 3a) a negative dip (–60%) is observed at ≈150 ps, which is a consequence of the fast emptying of the +1 polariton population due to the stimulated emission process: the –1 spin population exceeds the +1 and negative values of $\wp$ result. When this minimum is reached the stimulated emission process is over and the majority –1 polaritons will "slowly" flip their spin, bringing $\wp$ back to zero. Figure 3b shows that there is no negative $\wp$ for larger detuning, due to the modification of the stimulated emission dynamics.

In summary, our study of polariton dynamics as a function of exciton–cavity detuning and excitation density has revealed evidences of stimulated emission in microcavities. The coexistence of the two polariton branches for all excitation densities demonstrates that stimulated emission is polaritonic and due to the lower polariton branch. We have found a strong influence of exciton–cavity detuning on the spin relaxation of polaritons: the photoluminescence polarization degree reaches its maximum at a finite time and, for small normal mode splitting, negative values of $\wp$ appear.

Acknowledgements This work has been partially supported by "Fundación Ramón Areces", Spanish DGICYT under contract PB96-0085, the Spain–U.S. Joint Commission and the U.S. Army Research Office.

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