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Polaritonic coupling and spin dynamics in GaAs microcavities

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Abstract

We have used time-resolved photoluminescence spectroscopy to study the light emission dynamics in a semiconductor microcavity as a function of excitation density and exciton-cavity detuning. We paid special attention to polariton spin relaxation by using circularly polarized excitation. We have found a striking behavior of the photoluminescence degree of polarization, which reaches its maximum value at a finite time. As the excitation density is increased and the system enters the stimulated emission regime, this maximum is followed by a negative dip, whose depth strongly depends on exciton-cavity detuning. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

Microcavities are designed to enhance light–matter interaction. In the last decade, the optical studies on semiconductor microcavities have focused on both low and high excitation regimes. At low powers, the polariton dispersion [1,2], relaxation [3,4] and bleaching [5] have been investigated. At high excitation densities, non-linear properties of vertical cavity surface emitting lasers (VCSELs) and microcavities have been considered theoretically [6,7] and observation of laser-like emission has been reported [8–11]. Only recently, the spin properties of the carriers and their influence on light emission have been considered in VCSELs [6,8] and in microcavities [12]. The intermediate region has not been explored in detail. In this work, we report on the dependence of polariton relaxation and spin dynamics on excitation power.

The samples are grown by molecular beam epitaxy. They include dielectric mirrors separated by an $Al_{0.25}Ga_{0.75}As$ region in which three pairs of coupled GaAs quantum wells (QWs) are placed in the antinode positions of a $3\lambda/2$ planar microcavity. The interruption of the substrate's rotation during growth originates a slight wedge in the cavity thickness, which allows tuning the cavity resonance to the transition in the QW. A detailed description of the sample can be found elsewhere [12]. Our experiments were

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performed under non-resonant excitation using a conventional up-conversion spectrometer with a time resolution of ~2 ps. The excitation light was circularly polarized and the photoluminescence (PL) emitted by the sample was analyzed into its σ^+ and σ^- components by means of a $\lambda/4$ plate. The time evolution of the PL and of its degree of polarization, $\wp$, were studied as a function of excitation density and exciton–photon detuning.

2. Polariton recombination dynamics

Under low excitation density conditions, we have resolved a normal mode splitting ~300 ps after excitation. The splitting varies between 3.5 and 7 meV moving the excitation spot across the sample. This increase is due to the change in the photon-like mode energy caused by the variation of the cavity thickness. Taking into account the results of temperature dependent cw-PL and its magnetic field dependence [13], we have attributed an excitonic character to the lower polariton branch (LPB) and a photonic character to the upper polariton branch (UPB). We have confirmed that, under non-resonant excitation, there is no influence of the detuning on the PL's characteristic rise and decay times, in agreement with Ref. [14]. These times are similar for both polariton branches and amount to $\tau_r^X \sim 100$ ps, $\tau_d^X \sim 300$ ps, $\tau_r^\gamma \sim 70$ ps, $\tau_d^\gamma \sim 250$ ps, where $X(\gamma)$ and r(d) denote exciton-(photon-) like and rise (decay) time, respectively.

At high excitation densities the light emission dynamics changes drastically. Fig. 1 depicts a time-resolved PL spectrum taken 60 ps after excitation: the LPB appears as a very narrow peak (FWHM < 1 meV), together with a broader UPB (FWHM ~ 2 meV). Changing the detuning permits a control of their relative intensities. Fig. 2 shows the time evolution of the PL measured at the cavity mode energy for two different excitation densities. At low peak densities (~ 50 kW/cm^2, ●) the behavior is similar to that found, under non-resonant excitation, for excitons in QWs: the emission begins at zero time delay and is characterized by slow rise- and decay-times. The evolution is markedly different for high peak densities (~ 650 kW/cm^2, ○). There is an onset of the PL at ~30 ps, followed by a very fast rise and decay.[1] This finite time for the PL onset is related to the accumulation and relaxation of excitons, created above the cavity's stop band, towards energies where they can couple with the cavity.

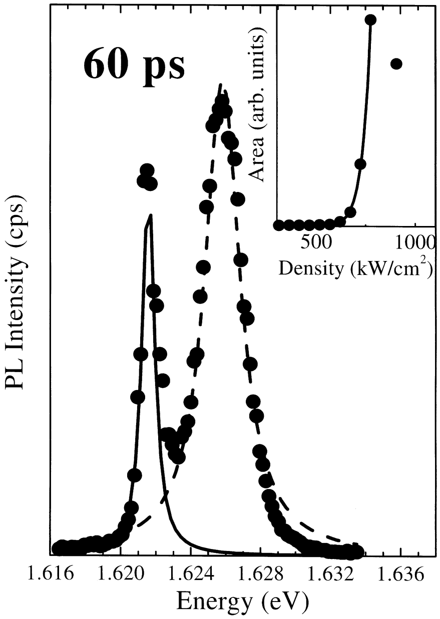

Fig. 1. PL spectrum 60 ps after excitation with ~ 500 kW/cm^2, $E_{\mathrm{exc}} = 1.706$ eV at 5 K. The inset shows the excitation density threshold of the integrated emission of the LPB.

The initial rise of the PL time evolution can be characterized by its curvature, η. The inset of Fig. 2 depicts η versus excitation density at 20 ps. A large increase is seen for peak excitation-densities above 500 kW/cm^2 (~ 20 W/cm^2, mean density). The integrated emission of the LPB presents a similar threshold density, as shown in the inset of Fig. 1. The threshold for both quantities increases with decreasing normal mode splitting. All these findings lead us to identify the narrow peak as the stimulated emission of the LPB.

Under high excitation conditions, the LPB is photon-like, while the UPB is exciton-like. Following the temporal evolution of both polariton branches, a clear anticrossing is observed once the stimulated emission is over [12]. At long times (> 500 ps) the LPB (UPB) recovers its exciton-like (photon-like) character. The stimulated emission has heavily

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[1] The peak at 110 ps is due to spin effects.

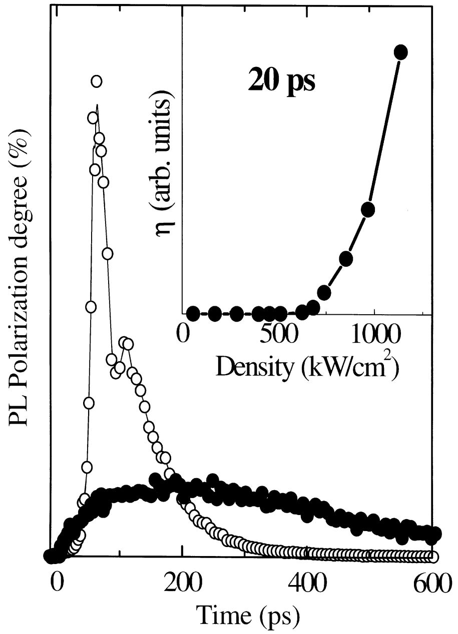

Fig. 2. Time evolution of the cavity mode for low (●) and high (○) excitation densities. Inset: PL's initial rise curvature as a function of excitation density.

decreased the polariton population and the emission spectra are now similar to those obtained under low excitation densities.

3. Polariton spin dynamics

In the following, we will call spin to the third component of the total angular momentum. An analysis of the temporal evolution of the PL's degree of polarization, $\wp$, gives direct information about the spin relaxation. Under σ^+ excitation, $\wp$ is defined as $(I^{\sigma^+} - I^{\sigma^-})/(I^{\sigma^+} + I^{\sigma^-})$ and is directly related to the difference between $+1$ and -1 spin populations. $I^{\sigma^\pm}$ denotes the PL emission with ± 1 helicity. In a QW, a σ^+ pulse will mainly create $+1$ excitons with center of mass momentum $\boldsymbol{K} \neq 0$. During their relaxation towards $\boldsymbol{K} = 0$ a -1 population will appear as a result of spin-flip processes. Eventually, both populations will be equal and therefore $\wp$ will vanish.

We have found that the time evolution of $\wp$ in a microcavity is very different to that typical of isolated QWs [15]. For bare excitons, $\wp$ reaches its maximum almost instantly after excitation. Then, spin relaxation processes will take $\wp$ back to zero. Let us concentrate first on the behavior of $\wp$ in microcavities at moderate excitation densities. Fig. 3 shows the time evolution of $\wp$ of the cavity mode for different excitation densities: $(*)$ and (○) below the threshold for stimulated emission and (◆) at threshold. The initial value of $\wp$ is only ~10% and $\wp_{\max}$ occurs a few tenths of ps after excitation. The value of $\wp_{\max}$ increases strongly with increasing excitation density, reaching values as high as 80%. The delay to reach $\wp_{\max}$ increases both with decreasing exciton-cavity detuning (not shown) and with increasing excitation density (Fig. 3). The fact that a finite time is needed to reach $\wp_{\max}$ implies that the exciton-cavity field interaction favors $+1$ polaritons at expenses of the -1 population. This process competes with the spin-relaxation, which tends to equalize both populations.

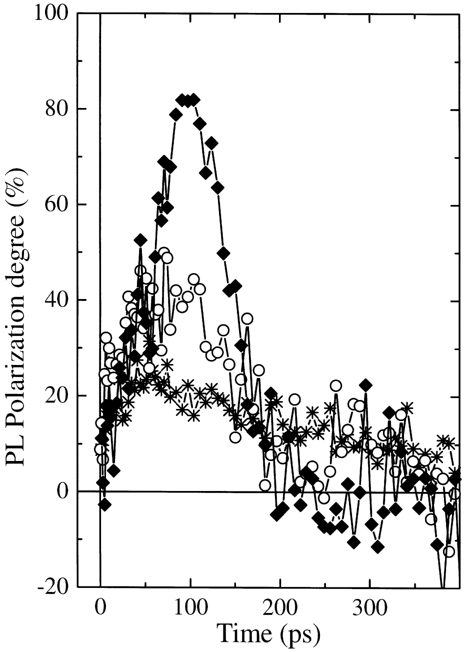

Fig. 3. Time evolution of the cavity mode PL's polarization for different excitation densities: $I_0(*), 2.3\,I_0$(○)$, 3\,I_0$ (◆).

Above the threshold for stimulated emission the time evolution of $\wp$ becomes even more conspicuous. Fig. 4 demonstrates that the $+1$ polaritons are initially 65% of the total population, i.e. $\wp \sim 30\%$, and

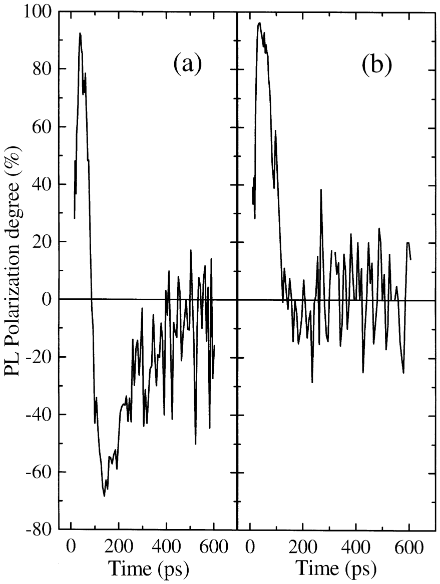

Fig. 4. Cavity mode PL's polarization degree measured at two different points of the sample, with small (a) and large (b) exciton-cavity detuning. The experimental conditions were the same for all the measurements.

they increase up to a 95% at ~30 ps.[2] The value of $\wp_{\max}$ does not depend on detuning. However, the temporal evolution of $\wp$, after the maximum is reached, is drastically modified by the detuning. For small normal mode splitting, Fig. 4a, a negative dip is observed (-60%), which is a consequence of the fast emptying of the $+1$ spin population due to the stimulated emission. The -1 polariton population exceeds the $+1$ and negative values of $\wp$ result. When the minimum value of $\wp$ is reached the stimulated emission is over, and the majority -1 polaritons will "slowly" relax their spin, bringing $\wp$ back to zero. Fig. 4b shows that, for larger normal mode splitting, the negative dip disappears due to modification of the stimulated emission dynamics by the exciton-cavity detuning.

In summary, we have found evidence for stimulated emission for the lower polariton branch in a microcavity. We have demonstrated the influence of detuning on the polariton dynamics. The PL degree of polarization has a maximum at finite times. This delay depends strongly on excitation density and normal mode splitting. A reversal of the polariton's spin is observed for small exciton-cavity detuning.

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[2] The same behavior is observed for the UPB, but the effect is much smaller ($\wp_{\max} \sim 60\%$).