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Spin-dependent coexistence of weakly coupled and strongly coupled modes in semiconductor microcavities

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Abstract

The dependence of the transition from the strong-coupling regime to the weak-coupling regime on the polariton spin orientation in a InGaAs semiconductor microcavity is experimental studied by means of time-resolved photoluminescence. Polaritons are created by non-resonant circularly-polarized optical excitation and the power intensity that breaks the strong coupling is found to be much lower for co-polarized polaritons than that for cross-polarized polaritons. Coulomb screening effects alone cannot explain the stronger loss of oscillator strength for majority excitons (co-polarized) and spin-dependent mechanisms are required to justify such behaviour.

Keywords: Microcavities; Polaritons; Ultrafast spectroscopy; Spin; Polarization

1. Introduction

In a quantum well (QW) an exciton is coupled to a continuum of photon modes. This mixed state of matter and radiation is called an exciton–polariton and it constitutes the true mode of propagation inside the semiconductor. The polaritonic effects are in general negligible, but when the QW is embedded in a microcavity with a cavity mode resonant with the excitonic transition,

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strong changes in the optical properties of the system can occur. If the exciton–photon interaction energy is greater than any homogeneous or inhomogeneous broadening of the bare photon or exciton mode, the system is in the strong-coupling (SC) regime. In the SC regime the polaritonic energy dispersion curves vs. $k_{/\!/}$ ($k_{/\!/}$ is the momentum in the QW plane) are strongly modified and the degeneracy of the exciton and photon level breaks into the lower polariton branch (LPB) and upper polariton branch (UPB), that are separated by a Rabi splitting Ω [1].

In a semiclassical model, the Rabi splitting is given by:

$$\Omega = 2\sqrt{|V|^2 - \frac{1}{4}(\gamma_c - \gamma)^2}$$

where V is the coupling parameter, proportional to the exciton oscillator strength, and γ_c and γ are the cavity linewidth and the non-radiative exciton linewidth, respectively.

When the density of carriers increases, the oscillator strength diminishes and the non-radiative linewidth broadens, resulting in a decreasing of the Rabi splitting [2]. As a consequence, when the excitation power increases, polaritons undergo a transition from the strong-coupling regime to the weak-coupling regime and the system emits at the bare cavity mode [3].

Here we will explore the spin dependence of the transition from the strong-coupling regime to the weak-coupling regime in an InGaAs semiconductor microcavity by means of time-resolved photoluminescence (PL) spectroscopy.

2. Sample and experimental setup

We study a $3/2\lambda$ microcavity of GaAs with a dielectric Bragg mirror of $Al_{0.1}Ga_{0.9}As/AlAs$ and two stacks of three $In_{0.06}Ga_{0.94}As$ QWs (100 Å) embedded at the anti-node position of the electric field in the cavity. The cavity mode energy is kept at resonance with the heavy hole exciton transition (detuning $\delta = E_{\text{cav}} - E_{\text{exc}} = 0$) at 5 K. Optical excitation is provided by pulses that are 2 ps long of a $Ti{:}Al_2O_3$ laser tuned at the energy of 1.63 eV, almost 180 meV higher then the considered exciton resonance. The electron–hole pairs excited at high energy create an exciton population with high momentum $k_{/\!/}$ that couples with the electromagnetic field and through phonon scattering processes relaxes towards the bottom of the polaritonic dispersion curve. PL from the $k_{/\!/} = 0$ state is selected by a pin hole and dispersed by a spectrograph that is coupled with a streak camera to allow both spectral and temporal analyses of the emission. A polarization optic is used to excite with circularly polarized light (σ^+) and distinguish the co-polarized (σ^+) and cross-polarized (σ^-) emission.

3. Results

In Fig. 1(a) and (b) are shown two streak camera images corresponding to σ^+ emission from $k_{/\!/} = 0$ under low and high excitation power P_{ex} ($P_{\text{ex}} = 0.1$ mW and $P_{\text{ex}} = 0.8$ mW, respectively). At $P_{\text{ex}} = 0.1$ mW the LPB and UPB are separated by a Rabi splitting $\Omega \approx 6$ meV and the long rise and decay times are typical of polaritons in the linear regime [4], while at $P_{\text{ex}} = 0.8$ mW a strong blue-shifted emission at short time, coexisting with the linear-regime emission, can be observed. At longer time the blue-shifted component recovers the linear-regime energy. In the figure, the cavity-mode energy (dashed line) and the energy of the linear and non-linear emission (arrows) are marked to highlight the diminished Rabi splitting at high power and the coexistence of two coupling regimes.

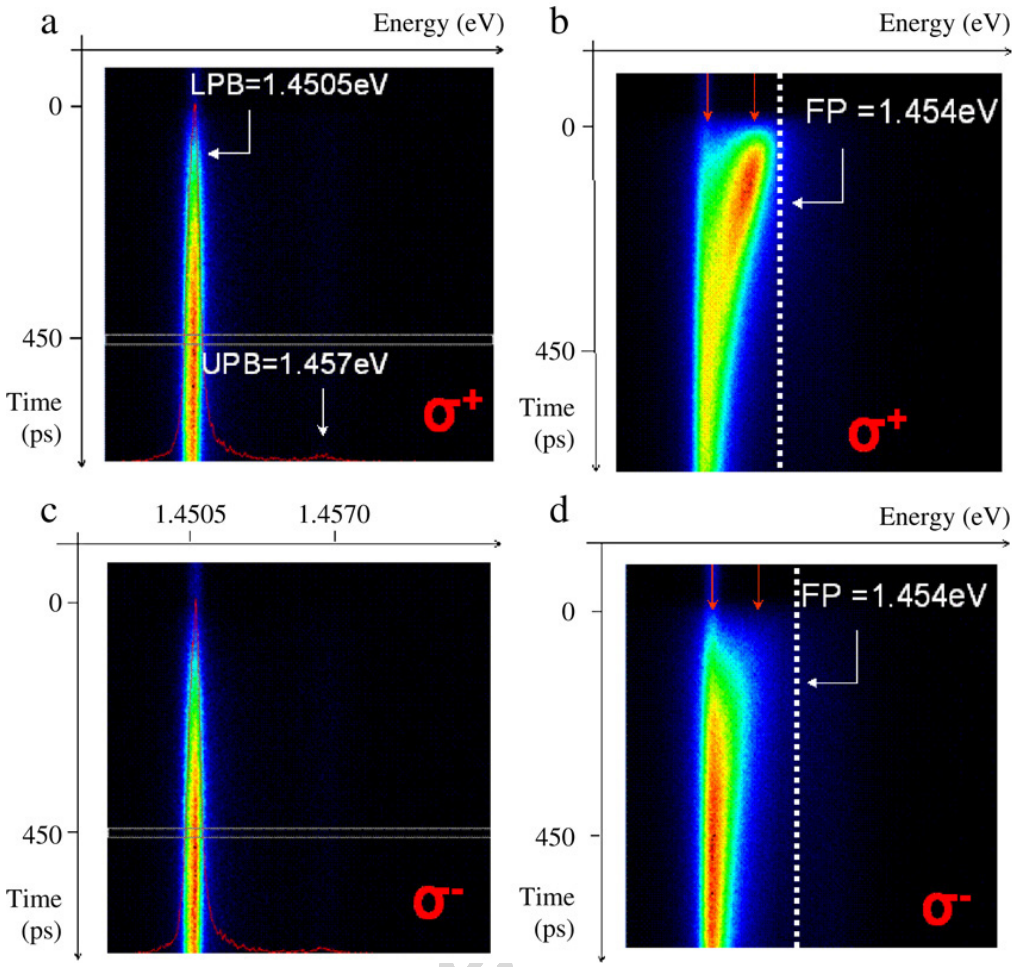

Fig. 1. Streak camera images of σ^+ emission for: (a) $P_{\text{ex}} = 0.1$ mW; (b) $P_{\text{ex}} = 0.8$ mW and σ^- emission for: (c) $P_{\text{ex}} = 0.1$ mW and (d) $P_{\text{ex}} = 0.8$ mW.

In Fig. 1(c) and (d) the streak camera images of σ^- polariton emission at the same excitation power $P_{\text{ex}} = 0.1$ mW and $P_{\text{ex}} = 0.8$ mW are presented. At low power, the traces corresponding to σ^+ and σ^- emission are very similar: the decay and rise times and the energies of the UPB and LPB are the same. The polarization degree, that compares the different emission intensities of the two polarizations, defined as $P = \frac{I_{\text{co}} - I_{\text{cross}}}{I_{\text{co}} + I_{\text{cross}}}$, is $P \approx 0$, typical for polaritons in the linear regime.

At $P = 0.8$ mW, a blue-shifted component broads the linewidth of the σ^- emission at short time, but the reduction of Rabi splitting is significantly weaker than in the σ^+ case (as marked by the arrows). The time-integrated PL intensities and polarization degree P are shown in Fig. 2: in this non-linear regime a large value of the polarization with a maximum $P \approx 50\%$ is obtained.

3.1. Coexistence of two dispersion curves

To check whether the coexistence of the linear and non-linear regime emissions at short times is due to the Gaussian profile of the laser beam ($d = 200$ μm), we have scanned the emission spot with a pin hole of $d = 10$ μm to analyze the PL coming from different positions of the excited area. In Fig. 3, the spectra observed with the pin hole placed at the center, and 50 μm and 100 μm from the center, respectively, are shown.

If the coexistence originated from a non-linear emission from the center and a linear emission from the border of the spot, where the intensity does not reach the non-linear threshold, a

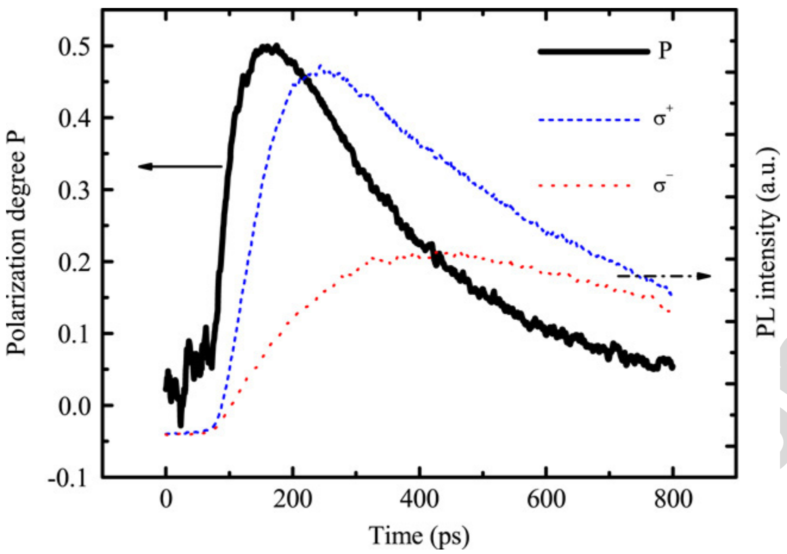

Fig. 2. Polarization degree P (solid line, left scale) and PL intensity for $P_{ex} = 0.8$ mW for the two polarizations σ^+ (dashed) and σ^- (dotted).

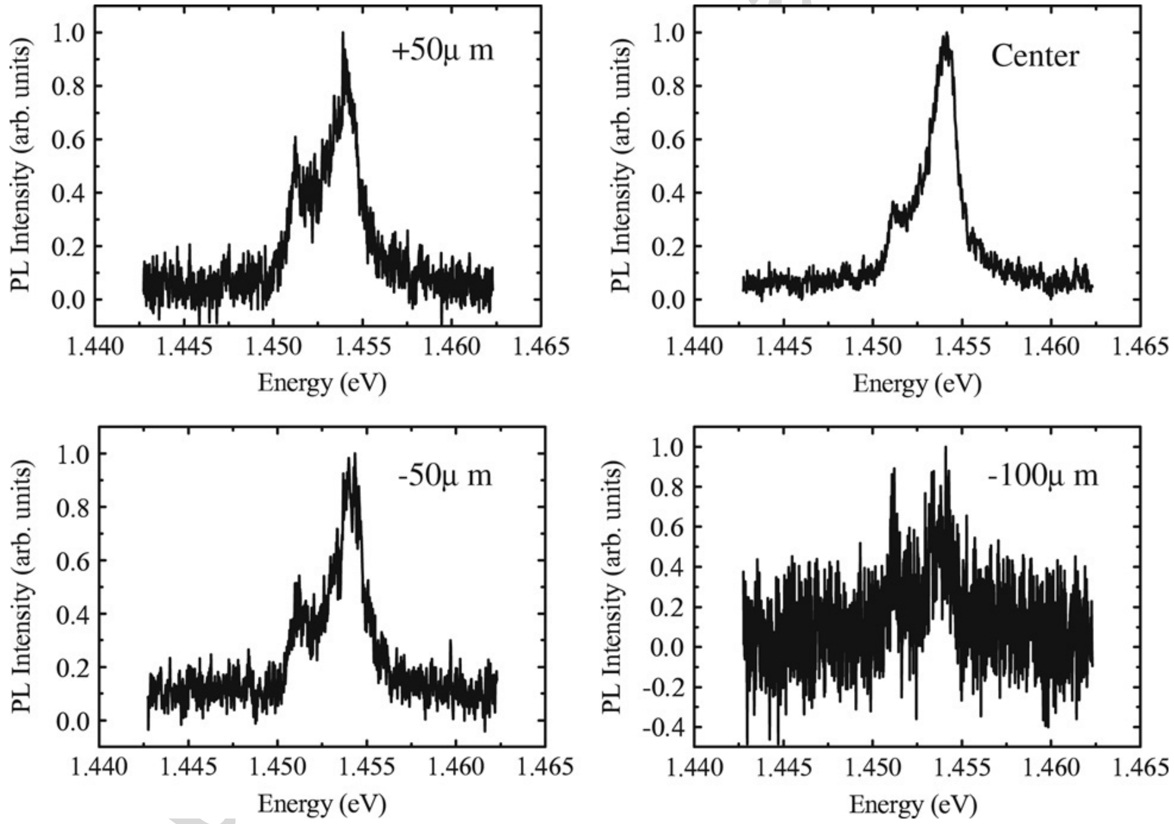

Fig. 3. PL spectra at different pin hole positions inside the excited area.

blue-shifted peak from the center and a lower energy peak from the border would be expected. However, we detect both components all over the excitation spot with our resolution of 10 μm. The peak intensity of both components presents a Gaussian profile along the excitation spot. Their intensity ratio is also well described by a Gaussian, though one could expect that the ratio would be superlinear on the excitation intensity, due to stimulated scattering process occurring where large excitation is present, and therefore not yielding a Gaussian profile. Moreover, the

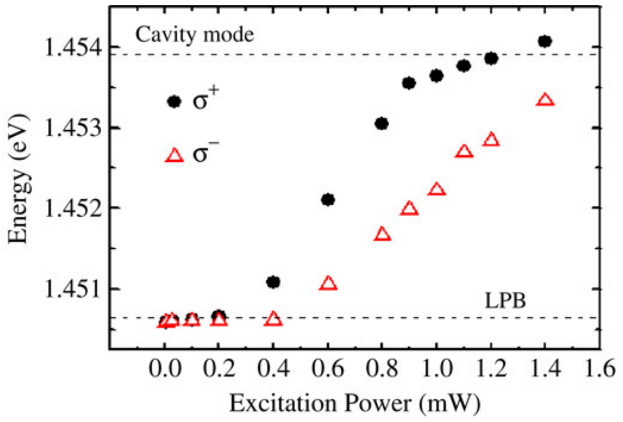

Fig. 4. LPB peak energies as a function of the excitation power for the two polarizations σ^+ (●) and σ^- (Δ).

separation in energy between the peaks remains constant over the whole emission area, ruling out the origin of the coexistence to the changes in the intensity profile of the excitation spot, which should yield a decreasing splitting moving from the center towards the edges of the spot. Linear and non linear-emission are clearly coexisting at every scanned position of the emission area and only the ratio of their intensities changes, indicating that the distribution of these regions, where the linear and non-linear emission originate and that are smaller than 10 µm, is following the Gaussian profile of the excitation spot.

3.2. Spin-dependent strong-coupling to weak-coupling transition

The LPB peak energies are displayed in Fig. 4 as a function of the excitation power for the two polarizations. It can be seen that at very low power intensity the emission energy is the same for σ^+ and σ^- polaritons, but a splitting appears at $P_{\text{ex}} = 0.4$ mW. Both polarizations undergo a loss of strong coupling with increasing intensity, but the Rabi splitting is different for σ^+ and σ^- polaritons under the same excitation conditions [5,6].

At $P_{\text{ex}} = 1$ mW, the σ^+ polaritons are in the weak-coupling regime (as can be readily inferred by its position that has reached the bare-cavity energy), while the σ^- polaritons' energy is still well below the cavity mode energy and a complete loss of the strong coupling is reached only at $P_{\text{ex}} = 1.7$ mW. The origin of this breaking of the spin degeneracy at high exciton density can be discussed in the light of the changes in the exciton oscillator strength. Two processes are likely responsible for these changes in a QW: the phase space filling (PSF), a blocking mechanism due to the exclusion principle, and the modification of the electron–hole (e–h) interaction induced by the presence of other e–h pairs [7]. Both exchange (also a consequence of the exclusion principle) and long-range Coulomb effects can modify the e–h interaction. In two-dimensional (2D) system, the effect of the long-range Coulomb effect can be disregarded [7].

To have a deeper understanding of the dependence of the energy splitting on the relative populations of opposite spin, we will look at the occupation factors of the σ^+ and σ^- polaritons. Selecting $k_{/\!/} = 0$ states by a pin hole, we can establish the number, N_k, of $k_{/\!/}$-states that contribute to the PL. With a calibrated measurement of the absolute power of the PL intensity it is then possible to estimate the number of polaritons, N_p, that are emitting with momentum $k_{/\!/} \sim 0$; therefore the occupation factor f_p, $f_p = N_p/N_k$, can be obtained [8].

In the case of $\delta = 0$, polaritons in the strong-coupling regime at the bottom of the LPB are a mixed state of excitons and photons, and the polariton occupation factor is calculated considering an exciton fraction of 50%. When the power is increased at $P_{\text{ex}} = 1$ mW, we consider the

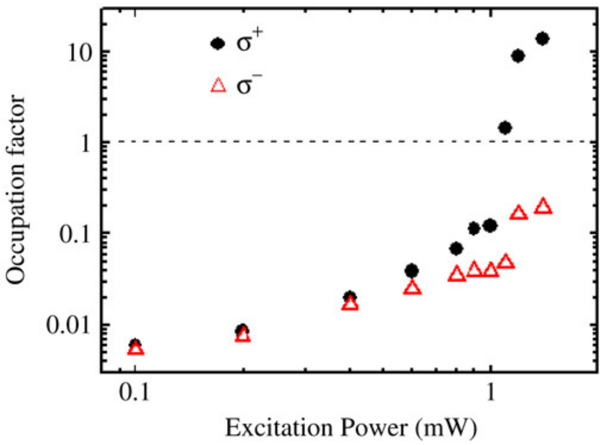

Fig. 5. Occupation factors as a function of the excitation power for the two polarizations σ^+ (•) and σ^- (Δ).

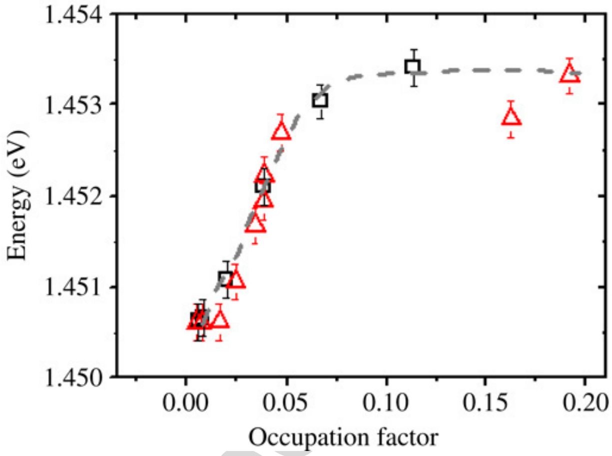

Fig. 6. LPB peak energy as a function of the occupation factors for the two polarizations σ^+ (•) and σ^- (Δ).

σ^+-state as 100% photonic while the σ^--state is still half-photon and half-exciton. In Fig. 5 the calculated occupation factors $f_p^{\sigma+}$ and $f_p^{\sigma-}$ are plotted as a function of P_{ex}.

Both $f_p^{\sigma+}$ and $f_p^{\sigma-}$ are smaller than one while the strong coupling dominates, hindering the possibility of stimulated polariton–polariton scattering [9]. The occupation factor $f_p^{\sigma+}$ reaches unity at $P_{\text{ex}} = 1.1$ mW, when the emission is fully photonic, and a laser-like emission (100% σ^+-polarized) is observed. Only at $P_{\text{ex}} = 2$ mW is a photonic laser emission from σ^- states obtained.

We will consider now the occupation factor of both polarizations only for the excitation powers at which polaritons are still in the strong-coupling regime. If we plot the σ^+ and σ^- LPB peak energies vs. the respective occupation factors as in Fig. 6, the two curves are exactly superimposed. Therefore one can conclude that the energies of emission of the polarized polaritons, and consequently the regime of existence of polaritons (strong vs. weak coupling), is fully determined by the occupation factor of the polaritons with a given spin and do not depend strongly on the total population of both kinds of polaritons.

This strong dependence of the emission energies on the relative populations of opposite spin has been explained for an exciton gas in a QW through an exciton–exciton interaction model [10]. According to this model the energy level splitting is due to many-body interexcitonic exchange while intraexcitonic exchange is responsible for the spin relaxation time. The spin level energy difference is directly proportional to the polarization [10], a fact that we have found also in our microcavity. However in QWs the σ^+ energy remains almost constant on increasing excitation intensity, and the splitting is mainly due to a red-shift of the σ^- emission energy, and the splitting vanishes, together with the degree of polarization, in a time of the order of 50 ps. In spite of the good qualitative agreement of the amount of energy splitting on the relative difference of the unbalanced populations, further investigations are needed to obtain a better understanding of this mechanism in a semiconductor microcavity. In fact, the dynamics of the emission is strongly modified by the cavity and the decreasing Rabi splitting effect on the polaritons energy has to be added in the calculations of the absolute position of the PL peaks.

4. Conclusions

We have studied the loss of the strong coupling with increasing excitation power for co-polarized and cross-polarized polaritons and obtained a significantly lower transition power for σ^+ polaritons than for σ^- polaritons. The loss of strong coupling for a polariton population of a given spin is determined by the occupation factor of that population and is not strongly influenced by the presence of opposite spin polaritons. The LPB spin degeneracy breaking is found to be directly proportional to the difference in the opposite spin populations. This spin-dependent strong-coupling to weak-coupling transition is probably due to a combined effect of the exciton resonance bleaching and exciton–exciton exchange interaction, but further investigations are needed to obtain a quantitative description of this effect.

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