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Polarization dynamics of microcavity polaritons: Three excitation regimes

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We present the results of time-resolved studies of polariton dynamics under non-resonant, circularly polarized excitation. We show how the emission intensity, its decay rate and the polarization behavior change in three characteristic excitation regimes and discuss the underlying relaxation mechanisms.

1 Introduction

Embedding a quantum well (QW) in a semiconductor microcavity enhances the interaction between the excitons and the photons. If the damping rates of the cavity photons and the QW excitons are smaller than their respective lifetimes, one deals with the strong coupling regime, where the interaction between the photons and the excitons leads to appearance of mixed states named cavity polaritons (for a review see e.g. [1]). The perspective of exploiting the polariton macroscopic coherence to build new devices, such as low threshold coherent light sources or optical parametric oscillators, lead to a large number of reports. Among others, the problem of spin dynamics of the cavity polaritons has been addressed. In particular, Martín et al. observed that after a non-resonant, σ^+-polarized excitation, the polarization dynamics of the polariton emission was strongly dependent on the detuning between the cavity and the bare exciton energies [2]. Moreover, they observed pronounced oscillations of the polarization of the emission perpendicular to the cavity surface, i.e. from the $k_{\|} = 0$ states. Subsequently, Kavokin et al. gave an explanation of the observed polarization oscillations within a pseudospin model taking into account the splitting between the TE and TM modes of the cavity [3]. They argued that linear polarization states, not circular ones, are the eigenstates of the system, and therefore due to their splitting, oscillations in the circular polarization are observed. However, their calculations predicted a degeneracy of the linear states at $k_{\|} = 0$. On the other hand, Martín et al. attributed the polarization to a splitting of circularly polarized $k_{\|} = 0$ polaritons, suggesting that these are the proper eigenstates at $k_{\|} = 0$ [2].

In this report, we present time-domain studies of the emission intensity and its circular polarization at a negative detuning in three characteristic excitation regimes.

2 Experiment

The sample under study was a λ-$Cd_{0.4}Mg_{0.6}Te$ microcavity with two QWs placed at both antinodes of the confined electromagnetic field. The top (bottom) cavity mirrors were distributed Bragg reflectors consisting of 17.5 (23) pairs of $Cd_{0.4}Mg_{0.6}Te/Cd_{0.75}Mn_{0.25}Te$ layers. The cavity was wedge-shaped, which

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allowed to vary the detuning between the bare exciton and cavity photon energies by changing the excitation spot on the sample. The Rabi splitting, evaluated from the detuning dependent emission studies, was equal to 10 meV. All the measurements were performed at a temperature of 5 K. The polariton emission was excited by 2 ps pulses from a Al_2O_3:Ti laser pumped with an Ar^{2+} ion laser. The excitation energy was tuned 62 meV above the lower polariton energy, i.e. to the first reflectivity minimum after the stop band. The excitation spot had a diameter of the order of 50 μm. The excitation beam was σ^+- or σ^--polarized and one circular polarization was detected to avoid the movement of the signal spot on the entrance slit of the monochromator. The signal was detected and time-resolved by a streak camera with an overall resolution of about 5 ps.

In this report, we concentrate on the results obtained for the emission from the $k_{\|} = 0$ at the detuning equal to -15 meV.

3 Results

In the time resolved emission spectra, three transitions are observed. The lower polariton (LP), the upper polariton (UP), and a third transition, located between LP and UP, which is attributed to the recombination of uncoupled excitons. The dependencies of the integrated emission intensity of these three transitions on excitation density (P) are presented in Fig. 1a. For LP, as P is initially increased, a quadratic intensity dependence is observed. As P is further increased, a threshold is reached at a density of about P_1 = 10 W/cm^2. Above this value, LP emission shows a dramatic increase until a second threshold is reached, at a density of approximately P_2 = 40 W/cm^2. Above the second threshold, the emission intensity increase with P becomes linear. In Fig. 1b, decay traces of the LP emission for four excitation densities are presented. For densities below P_1, the decay of the LP emission is slow: the decay time is of the order of 200 ps. Above P_1 a substantial acceleration is observed: the decay times become as short as 20 ps. Above P_2 no further change is recorded.

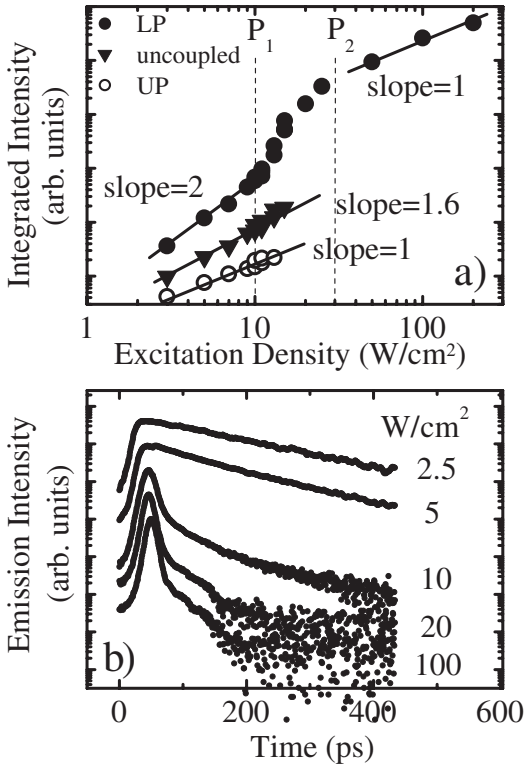

Fig. 1 a) Integrated intensity of the LP (full circles), UP (empty circles), and uncoupled exciton (triangles) emissions as at a detuning of -15 meV and $k_{\|} = 0$ as function of the excitation density. b) Decay traces of the LP emission for various excitation densities.

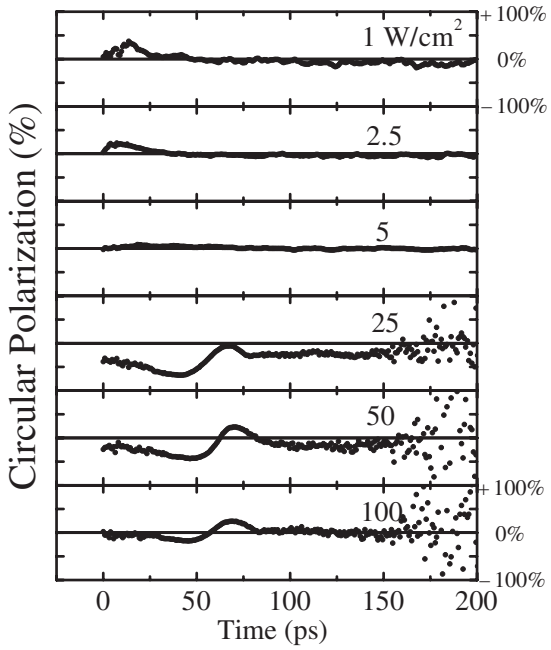

Fig. 2 Temporal profiles of circular polarization of the lower polariton emission for various excitation densities at a detuning of -15 meV and $k_{\|} = 0$.

We define the polarization as $P = \dfrac{I^{\mathrm{co}} - I^{\mathrm{cross}}}{I^{\mathrm{co}} + I^{\mathrm{cross}}}$, where I^{co} and I^{cross} are the emission intensities co- and cross-polarized with respect to the excitation beam, respectively. The temporal profiles of the polarization for various excitation densities are presented in Fig. 2. Below P_1, we observe a positive polarization of the order of 20%, which decays within about 25 ps. The maximum value of this polarization decreases with increasing excitation density and virtually disappears at P_1. As P is further increased, at short delays after the excitation, the polarization has negative values and then exhibits oscillations with a period of about 30 ps. The peak to peak change in the polarization due to these oscillations amounts to roughly 80%. As the excitation density is increased above P_2, the oscillations remain, but the peak to peak variation decreases to about 40%.

4 Discussion

After a non-resonant excitation pulse, photocreated electron–hole pairs loose most of their momentum via scattering with LO phonons. Large $k_{\|}$ exciton-like polaritons are formed, which then relax down the dispersion curve. For negative detunings, the lower polariton branch has a large photonic character. Therefore, approaching $k_{\|} = 0$, the curvature of the dispersion becomes very large and the density of states (proportional to the polariton mass) very small. Consequently, phonon scattering, as an energy relaxation mechanism, which would lead to a linear dependence of emission intensity on P, becomes inefficient, and a large-$k_{\|}$ reservoir of polaritons is formed [4, 5]. The energy relaxation from the reservoir to $k_{\|} = 0$ states occurs via spontaneous polariton–polariton scattering, resulting in a quadratic dependence of the emission from $k_{\|} = 0$ on P [6–8]. Therefore, we denote the excitation density regime below P_1 as spontaneous regime. This relaxation bottleneck effect disappears as the number of polaritons is further increased. Above a certain density, polariton–polariton scattering becomes stimulated by the occupation of the final $k_{\|} = 0$ state. This effect results in a significant acceleration of the relaxation processes [9] and, consequently, in an exponential increase of the emission intensity with increasing excitation density [10]. The regime between P_1 and P_2 is the stimulated regime. A subsequent increase of the polariton population results in bleaching of the exciton transition due to exchange interaction and phase space filling [11]; the bosonic description ceases to be valid, and a transition to weak coupling regime takes place.

Indeed, above P_2, in is the saturation regime, the transition energy becomes equal to the energy of the bare cavity mode.

The power dependence of UP emission intensity remains linear in the spontaneous regime, which points out that the UP states are populated via phonon scattering. The previously proposed process in which two reservoir polaritons scatter into LP and UP states [12] is therefore inefficient in our system due to a small density of final states. On the other hand, the uncoupled exciton transition exhibits a superlinear dependence on excitation power. This result suggests that more than one particle is involved in the scattering into this state.

It is worth noting that the occurrence of one of the three excitation regimes described above is not strictly dependent on the density of initially excited particles, but on their transient density in the microcavity. As the polariton (or an electron–hole pair, in the case of saturation regime) population decreases, the system undergoes a transition from one regime to another in time [9]. Indeed, as seen in the temporal trace of the LP emission under excitation with 10 W/cm^2, corresponding to the onset of the stimulation regime (Fig. 1b), at short delays after the excitation, the decay is very fast showing the characteristics of the stimulated scattering. However, after 100 ps, the recombination slows down and the decay occurs with a time constant characteristic of the linear regime. After the rapid initial decay, the $k_{\|} = 0$ states are emptied and the final state stimulation does not take place any longer. Consequently, the recombination slows down, and the systems returns to the spontaneous regime. An analogous effect is seen in the energy positions of the transitions observed in the saturation regime. Figure 3a presents these energy values, obtained from fitting a Lorentzian to the transient spectra, versus time for co- and cross-polarized emissions, together with the total emission intensity. At delays shorter than 50 ps, the system is in the saturation and weak coupling regime, and the transition energies are equal to the bare cavity mode energy. As the emission intensity decreases, the system clearly moves to strong coupling and stimulation regime.

In Fig. 3c the integrated circular polarization is depicted. In the spontaneous regime, the positive polarization is due to the transfer of electron and hole optical orientation to the polariton $k_{\|} = 0$ state. In the

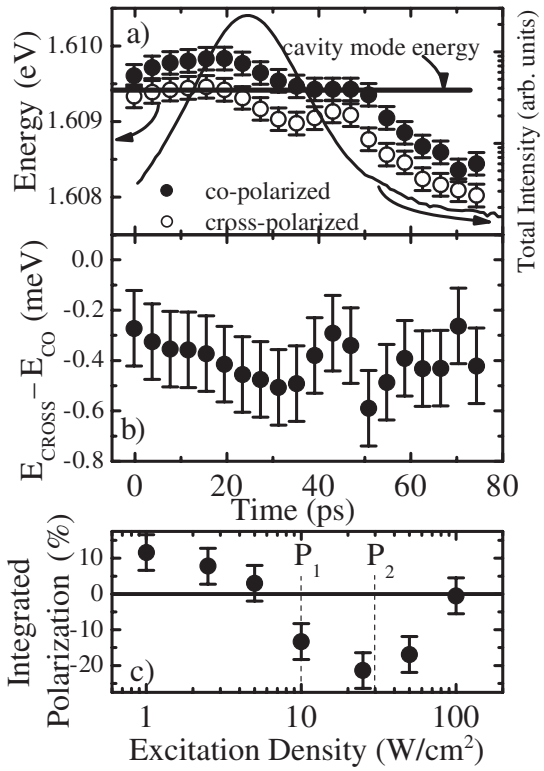

Fig. 3 a) Energy positions of co- (full) and cross-polarized (empty) LP transitions, obtained from fitting a Lorentzian, together with total (co + cross) emission intensity for an excitation density of 100 W/cm^2. A transition from weak to strong coupling is observed at delays of about 50 ps after the excitation pulse. b) Splitting between cross- and copolarized LP transitions calculated from the results of a). c) Time-integrated polarization as a function of the excitation density.

stimulated regime, a negative polarization value is obtained indicating that certain large-$k_{\|}$ states have to emit light positively polarized in order to conserve the total angular momentum [13]. In the saturation regime, the oscillations give integrated zero polarization (see Fig. 2). As can be seen from Fig 3b, there is a splitting between the cross- and co-polarized components. A possible origin of this splitting is the interexcitonic exchange interaction. It has been observed, that the spin degeneracy of QW excitons is broken when a large majority spin population is present in a QW [14]. The energy of the minority spin excitons is decreased and the resulting splitting is directly proportional to the difference between majority and minority spin populations. In the case of microcavity polaritons, this splitting has a complicated dependence on both $k_{\|}$ and time, that has to be considered in a full description of the polarization oscillations. In particular, this splitting should produce oscillations also in the linear polarization degree under circular pumping.

In summary, we presented an analysis of the dynamics of light emission from a semiconductor microcavity. We found that there are three characteristic excitation regimes, which determine the dynamics of the emission intensity and its circular polarization degree. In the spontaneous regime, the relaxation toward $k_{\|} = 0$ takes place via polariton–polariton pair scattering, and the polarization originates from the optical orientation of photoexcited carriers. In the stimulated regime, the relaxation to $k_{\|} = 0$ states is substantially enhanced due to bosonic final state stimulation. In this regime, polarization oscillations are developed due to splitting between linear TE and TM modes of the cavity. In the saturation regime, the strong coupling between the QW excitons and cavity photons is lost, and a transition to electron–hole plasma lasing is observed. In this regime, the polarization oscillations are still observed, however their amplitud is decreased.

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References

[1] Special Issue on Semiconductor Microcavities, edited by J. J. Baumberg and L. Viña, Semicond. Sci. Technol. **18** (2003).
[2] M. D. Martin, G. Aichmayr, L. Vina, and R André, Phys. Rev. Lett. **89**, 77402 (2002).
[3] K. V. Kavokin, I. A. Shelykh, A. V. Kavokin, G. Malpuech, and P. Bigenwald, Phys. Rev. Lett. **92**, 17401 (2004).
[4] F. Tassone, C. Piermarocchi, V. Savona, and A. Quattropani, P. Schwendimann, Phys. Rev. B **56**, 7554 (1997).
[5] M. Müller, J. Bleuse, and R. André, Phys. Rev. B **62**, 16886 (2000).
[6] P. Senellart, J. Bloch, B. Sermage, and J. Y. Marzin, Phys. Rev. B **62**, R16 263 (2000).
[7] P. G. Savvidis, J. J. Baumberg, D. Porras, D. M. Whitaker, M. S. Skolnick, and J. S. Roberts, Phys. Rev. B **65**, 073309 (2002).
[8] D. Porras, C. Ciuti, J. J. Baumberg, and C. Tejedor, Phys. Rev. B **66**, 085304 (2002).
[9] M. D. Martin, L. Viña, J. K. Son, and E. E. Mendez, Solid State Commun. **117**, 267 (2001).
M. D. Martin, L. Viña, and E. E. Mendez, Solid State Commun. **119**, 259 (2001).
[10] Le Si Dang, D. Heger, R. André, F. Bœuf, and R. Romestain, Phys. Rev. Lett. **81**, 3920 (1998).
[11] S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, Phys. Rev. B **32**, 6601 (1988).
[12] R. Huang, F. Tassone, and Y. Yamamoto, Phys. Rev. B **61**, R7854 (2000).
[13] L. Viña, R. André, V. Ciulin, J. D. Ganiére, and B. Deveaud, Inst. Phys. Conf. Ser. **171**, G4_2 (2003), edited by A. R. Long and J. H. Davies (Inst. of Physics Publishing, Bristol, UK).
[14] L. Viña, E. Muñoz, E. Pérez, J. Fernández-Rossier, C. Tejedor, and K. Ploog, Phys. Rev. B **54**, R8317 (1996).