Physics of Semiconductors 2002. Institute of Physics Conference Series 171, G4_2 (2003), ed. by A R Long and J H Davies (Institute of Physics Publishing, Bristol, UK)

Time- and angle-resolved emission of a microcavity in the non-linear regime

L. Viña[1], R. André[2], V. Ciulin[3], J.D. Ganiere[3] and B. Deveaud[3]

[1]Dept. Física de Materiales, Universidad Autónoma, E-28049 Madrid, SPAIN

[2]Lab. Spectrométrie Physique (CNRS), Univ. Joseph Fourier 1, F-38402 Grenoble, France

[3]Physics Dept., Swiss Federal Institute of Technology, CH-1015 Lausanne, Switzerland

Abstract. We have observed strong oscillations in the time-resolved emission of a semiconductor microcavity, excited non-resonantly with circularly-polarized light pulses. When the cavity is optically driven into a non-linear emission regime, the period of the oscillations amounts to ~30 ps, independently of the angle of observation. The angular dependence of the emission, at a negative detuning of -13 meV, shows that although the cavity starts emitting with maximum intensity at $\mathbf{K}$~0 cm^{-1}, the intensity is rapidly transferred to $\mathbf{K}$~2x10^4 cm^{-1}, close to the inflection point of the lower polariton branch, giving rise to a ring emission in a cone centred at ~15º. At small angles the polarization of the emitted light reverses its helicity with respect to that of the exciting pulses and steadily becomes positive for large angles.

1. Introduction

The study of the optical properties of semiconductor microcavities driven optically in a non-linear response regime has flourished in the last years due to the possibility of obtaining a polariton condensate. [1-3] The largest effort has been devoted to the investigation of the strong coupling regime, in which the eigenstates of the system are not longer pure exciton or pure photon but a superposition of both, known as cavity polaritons [4]. The strong mixing between excitons and photons modify strongly the in-plane dispersion relations of the polaritons [5]; this modification is easily observable by changing the angle of detection in optical experiments [6].The reduction of the density of states and the bosonic nature of the polaritons leads to the possibility of stimulated scattering effects at densities well below the exciton saturation density. These effects have been observed experimentally in III-V and II-VI microcavities [7-13]. More recently, it has been shown that in III-V microcavities stimulated polariton scattering cannot be achieved under high-energy non-resonant excitation conditions [14], because saturation of the strong coupling regime takes place before an occupation factor of the polaritons close to unity is obtained. However, in II-VI microcavities, and in particular in

CdTe based materials, the existence of stimulated scattering and probe amplification under non-resonant conditions seems to be well established [15,16].

On the other hand, a renewed interest on the carriers' spin in semiconductor structures has given rise to a new field, *spintronics*, which explores the possibility of designing new spin-based devices. In the particular case of cavity polaritons, due to the mixed photon-exciton character, significant changes on their spin dynamics with respect to bare quantum wells are expected. Nevertheless, the spin has been considered only very recently both under cw [17] and pulsed excitation [10,18-22]. In this work we present new results on the polariton dynamics, with special emphasis on its spin behavior, in II-VI microcavities under non-resonant conditions in the strong coupling regime.

2. Experimental details

The sample is a $Cd_{0.40}Mg_{0.60}Te$ microcavity of λ thickness, sandwiched between the top (bottom) distributed Bragg reflectors (DBRs). These mirrors are made of 17.5 (23) pairs, of alternating $\lambda/4$-thick layers of $Cd_{0.40}Mg_{0.60}Te/Cd_{0.75}Mn_{0.25}Te$. Two pairs of 90-Å thick CdTe quantum wells are placed at the antinodes of the cavity to obtain the optimum radiation-matter interaction, which leads to a Rabi splitting of ~ 14 meV. A slight wedge in the cavity thickness allows tuning the cavity and the exciton into resonance by moving the excitation spot across the wafer.

The microcavity is mounted in an immersion cryostat, where its temperature is kept at 2 K, and is optically excited with 1.5 ps pulses at the first minimum above the stop-band of the DBRs. The emission, angle-resolved using a small pinhole, which obtains a resolution of 1º, is time- and spectrally-resolved using a streak camera with a time resolution of ~ 5 ps. For polarisation-resolved measurements, a pair of $\lambda/4$ plates are included in the experiment: the excitation light is σ^+-polarised and the PL emission is analysed into its σ^+- and σ^--polarised components. All the experiments presented here are performed at a negative detuning of -13 meV with power densities below 50 W cm^{-2}, which ensures that the strong-coupling regime is maintained.

3. Results and discussion

Figure 1 shows the time evolution of the photoluminescence, detected at the lower polariton branch (1.61 eV), exciting at the first minimum above the stop-band (1.686 eV), for different angles of detection ranging from 0º to 23.5º, for a negative detuning of δ= -13 meV and an excitation density of 45 W cm^{-2}. Under these conditions, the strong coupling regime is still preserved as can be inferred by the energy position of the emission from the lower polariton branch (not shown), which at resonance, δ=0, has shifted only 2 meV towards the blue, without collapsing the Rabi splitting. The excitation pulses where right-handed circularly polarized (σ^+) and the emission was analyzed into its σ^+-component. Several aspects are worthwhile to be discussed in the dynamical behaviour of the light emission:

a) The strongest emission occurs at an angle of ~15º and not at 0º.
b) Striking periodic oscillations of the emission intensity are obtained, with a period independent of the observation angle.
c) The time-delay to reach the first maximum of the emission increases monotonically with increasing angle.

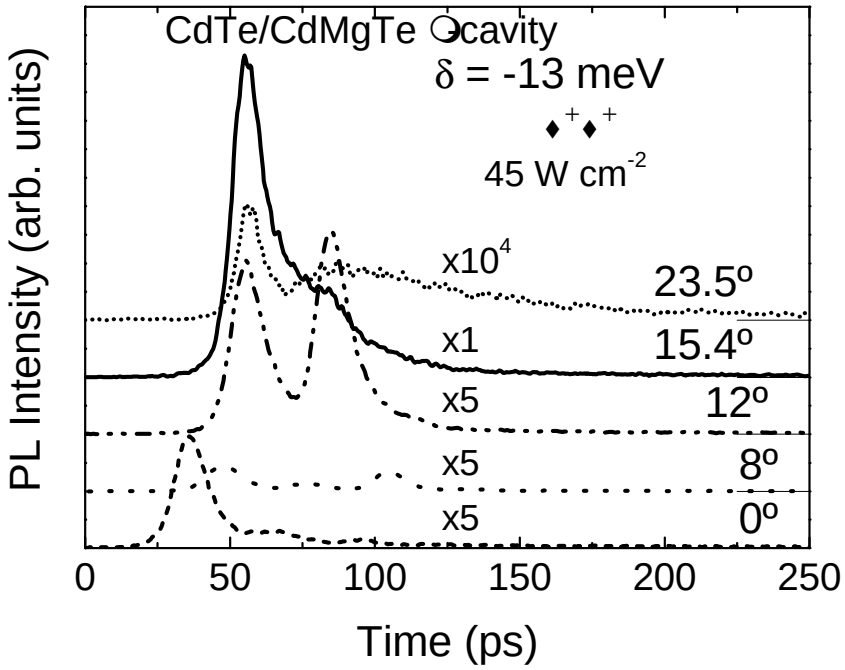

Figure 1. Time traces of the CdTe/$Cd_{0.4}Mg_{0.6}$ Te microcavity emission for different angles. The data are taken at a negative detuning of –13 meV, exciting with σ^+-polarized pulses at the first minimum of the stop band with a power density of 45 W cm^{-2}, and detecting the σ^+ emission. The traces are enlarged by the factor shown in the figure. Note the oscillatory behaviour with a period of 30 ps.

The non-resonant excitation of the sample creates excitons that rapidly relax into polaritons in the cavity. The emission starts at **K**~0 (0º), at these early times the polaritons are confined into an energy trap and the dynamics of the light emission is governed by final-state stimulated scattering, which is responsible for the very fast rise- and decay-times of the PL. The bosonic character of the final-state stimulated scattering is evidenced by an exponential increase of the PL intensity from the photon-like lower polariton branch with excitation power, similar to that observed in pump-and-probe [10] and cw- photoluminescence experiments [11]. This evidences a self-stimulated emission from the **K**~0 states after excitation of very large **K** excitons, which relax to finite momentum polaritons in the lower branch. These polariton distribution undergoes polariton-polariton scattering, and for a magic angle [10] one of the particles ends up at **K**~0. However, this behaviour is transient and at later times the maximum intensity of the emission occurs at an angle of 15.4º (**K**~$2x10^4$ cm^{-1}), giving rise to a ring emission, which has been also observed in resonant- excitation cw [13,23] and time-resolved experiments [24].

Figure 2 depicts the dispersion of the lower polariton branch (open circles) for the same conditions shown in Fig.1, together with the dependence of the normalized integrated intensity on the observation angle (triangles). It is clearly seen that the maximum intensity is obtained at 15º, close to the inflection point of the lower polariton branch. The intensity decays abruptly at larges angles and more smoothly at smaller angles, reaching a minimum at ~7º and increasing again slightly at 0º, reaching a value one order of magnitude smaller than that at 15º.

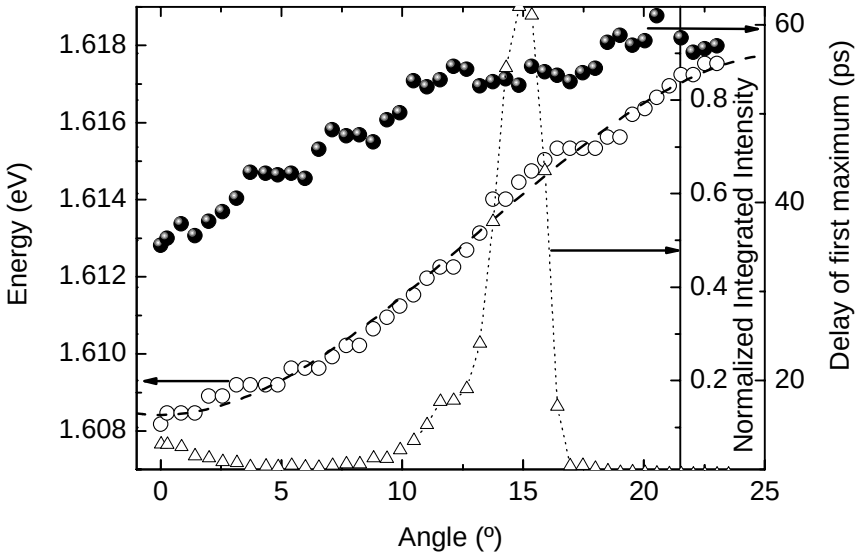

Figure 2. Dispersion of the lower polariton branch (open circles, left scale) of the CdTe/$Cd_{0.4}Mg_{0.6}$ Te microcavity. Normalized integrated intensity (triangles, medium right scale) and time-delay of the first maximum in the emission (solid circles, right-most scale) as a function of observation angle for δ= -13 meV and a power density of 45 W cm^{-2}.

The solid circles in Fig. 2 represent the temporal delay needed to reach the first maximum in the emission intensity (see Fig.1). This delay increases monotonically with the observation angle, saturating for large angles, when the lower polariton branch becomes increasingly more excitonic-like. This behaviour evidences the dynamical appearance of the ring emission, which can be related to the time evolution of the polariton population distribution and to the existence of a bottleneck in the rate of energy relaxation of the excitons into the **K**~0 polariton trap. At short times (but long enough to allow the non-resonantly created excitons to form a polariton distribution), the polariton population is the highest along all its dispersion and the emission originates exclusively from **K**~0 states (this occurs before a large number of polaritons escape from the microcavity). This resembles the situation obtained in cw experiments where strong coherent emission is observed simultaneously with a ring above a threshold excitation density [23]. In these experiments is also reported that at low excitation powers the diameter of the ring emission increases, what is equivalent to our observation that at longer times after the excitation, when the polariton population has decreased considerably, the emission shifts to large **K** vectors and peaks at **K**~$2x10^4$ cm^{-1} (~15º). This distribution is rather sharp with a FWHM of 2.5º and it is responsible for most of the emission of the microcavity even at large excitation powers, when the system is driven in a non-linear regime. It has been proposed that the existence of this non-thermal polariton distribution could help the start of stimulated scattering towards **K**~0 states [6], but we observe that the emission at **K**~0 happens before this large **K** distribution is formed. Our results demonstrate the importance of a temporal- and angular-resolved analysis of the emission in order to understand more deeply the complex behaviour of the polariton distribution in microcavities under high-excitation conditions. The difference in energy of 6 meV between the emission at 0º and 15º accounts also for a shift reported in Ref. 16 where a non-linear cw-emission is obtained for large excitation powers and the cavity is still in the strong coupling regime.

The excitation intensity dependence of the emission at 15º also reveals a process of stimulation at **K**~2x10^4 cm^{-1}. Our interpretation coincides with the one proposed by Alexandrou *et al*. [16]: the excitation pulse creates electron-hole pairs in the continuum of the quantum well embedded in the cavity, which after scattering by LO phonons form large **K** excitons. Two excitons with **K** of different sign may scatter, via a Coulomb mediated mechanism, producing two polaritons in the upper- and lower-polariton branch, respectively, conserving energy and momentum. If the population of the lower-polariton state reaches unity, final-state stimulated scattering is obtained, which lasts as long as there is sufficient population in the exciton reservoir.

More conspicuous is the observation of strong oscillations in the emission intensity, with a period of 30 ps, which is independent of the observation angle and of the helicity of the emitted light (see Fig. 3 below). The trace corresponding to an angle of 12º in Fig. 1 reveals that the intensity becomes stronger, for certain angles, after the first period, a behaviour not reported before, to the best of our knowledge, in any time-resolved emission of a semiconductor heterostructure. The energy scale corresponding to the beat period is of 136 μeV, which does not resemble any characteristic energy of the microcavity. A similar beat period, described recently for wire-shaped microcavities [25], has been attributed to uniaxial strain in the quantum well plane resulting from the cavity patterning to form wires. The strain release causes a reduction of the quantum well symmetry, which is assumed to be responsible for a lifting of the exciton degeneracy, due to exchange interaction. The corresponding splitting is also observed, its value being compatible with the beat period, and showing an oscillatory behaviour. Indeed, the beats reported for the light intensity and energy of the emission are in antiphase for σ^+ and σ^- polarization [25].

Figures 3a and 3c demonstrate that in our case the beats are in phase for both orthogonal polarizations and that the period is independent of the detection angle. The existence of strain as responsible for an energy splitting and the resulting beats in the time domain can, in principle, be excluded in our system. A possible explanation for the existence of the beats is a manybody effect, as a result of exchange and vertex corrections to the self-energies of dense exciton gases [20,21,26]. However, within the spectral resolution of our experiments we are unable to detect an energy splitting of the order of 136 μeV. This splitting would be, on the other hand, compatible with the observed values of the degree of polarization obtained in our experiments [26].

Let us now turn into the discussion of the spin dynamics of the polaritons. Figure 3a evidences that at **K**~0, after excitation with a σ^+-polarized pulse, the counter-polarized σ^--emission (dotted line) is stronger than the co-polarized emission (solid line). This fact results in a negative degree of polarization, defined as

$$\tilde{A} = \frac{I^{++} - I^{+-}}{I^{++} + I^{+-}}$$

with I^{++} (I^{+-}) the emission intensity exciting with σ^+ polarization and detecting the $\sigma^+(\sigma^-)$ component, as clearly seen in Figure 3b. The polarization degree reaches a minimum value of -30 % before decaying rapidly back to zero.

A detailed discussion of the polarization behaviour can be found in Ref. 21. In the microcavity discussed in this paper the absolute values of the negative polarization at 0º (integrated $\wp$ = -25 %) are smaller than those reported in Ref. 21 due to different conditions of the experiments such as: sample geometry, Rabi-splitting [19], detuning and excitation power density in the sample. The absolute values of the polarization decrease monotonically with increasing angle up to 5º, where the polarization becomes zero. A further increase of the observation angle leads to positive values of $\wp$,

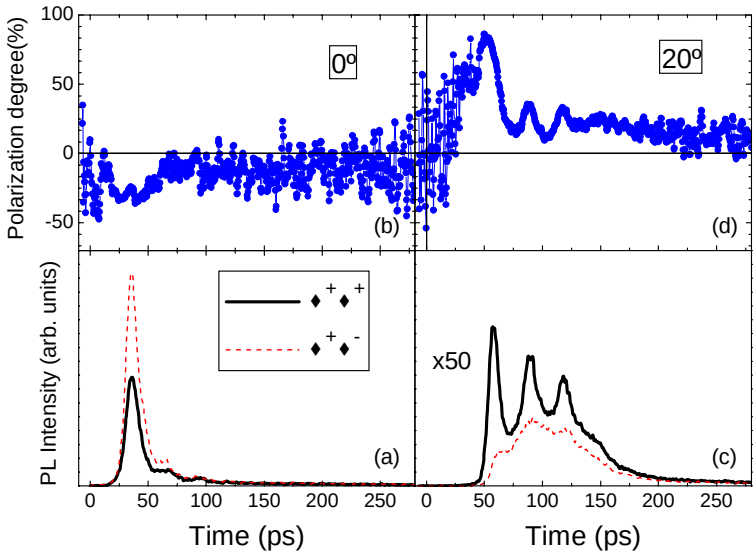

Figure 3. (a) Time evolution of the CdTe/$Cd_{0.4}Mg_{0.6}$ Te microcavity emission at 0º, analysed into its σ^+-(solid line) and σ^--components (dotted line). (b) Time evolution of the degree of polarization for 0º. Panels (c), with traces enhanced by a factor of 50, and (d) are equivalent to (a) and (b), respectively, detecting at 20º. The data are taken at δ= –13 meV, exciting with σ^+-polarized pulses at the first minimum of the stop band with a power density of 45 W cm^{-2}.

which keeps increasing attaining an integrated value of +20 % at 15º and saturating for larger angles. Figure 3c depicts the time evolution for both orthogonal polarizations of the emitted light at a detection angle of 20º, the intensity has decreased considerably (~50 times) with respect to that at zero angle. The beats in the σ^+- and σ^-- emission are clearly seen and now the co-polarized component (σ^+) is definitively larger than the counter-polarized emission. This is clearly reflected in the polarization shown in Fig. 3d, which has now positive values at all times. The integrated value of $\wp$ is 20%, a peak (80%) in its time evolution is obtained at finite times (~50 ps), as it was also the case for negative values of polarizations, in contrast with the situation in bare quantum wells where the polarization always decays monotonically from its value at zero time-delay [19]. The beats observed in the emission are also clearly seen in the time evolution of $\wp$. It is worthwhile to mention that oscillations in the time evolution of the polarization have been also hinted in the non-linear emission of GaAs microcavities [19] and in up-conversion experiments of CdTe microcavities [21] but the high noise level hindered an unambiguous determination of their existence.

The fact that most of the emission of the cavity, i.e. for angles larger than 5º, has a positive degree of polarization ($I^{+\,+} > I^{+\,-}$) is in agreement with the conservation of angular momentum, and validate the results obtained in the up-conversion experiments [20,21], where the surprising observation of very large negative values of the polarization was made. These negative values apparently could contradict the conservation of angular momentum, but one has to take into account that with the up-conversion technique only emission close to 0º was detected. Since the excitation with σ^+-polarized pulses was also non-resonantly, a larger population of excitons with J_z =+1 are created, but -1 excitons are also present. The latter suffer stimulated scattering to **K**~0 [20,21], giving rise to the negative values of polarization.

5. Summary

Strong oscillations in the time-, polarization- and angle-resolved light emission of a CdTe microcavity are observed when the system is optically driven into the non-linear regime. The oscillations are in phase for right- and left-handed helicity of the emitted light and its period amounts to 30 ps independently of the observation angle. The emission starts at **K**~0 cm^{-1} (0º) with negative values of the polarization and rapidly evolves into a ring emission centred at 15º (**K**~2x10^4 cm^{-1}), which present positive values of the degree of polarization.

Acknowledgments

This work has been partially supported by the EU (TMR-Ultrafast Quantum Optoelectronics Network), the Spanish DGICYT (PB96-0085) and the CAM (07N/0064/2001). We thank D. Porras and C. Tejedor for helpful discussions.

References

[1] Tassone F, Piermarocchi C, Savona V, Quattropani A and Schwendimann P 1996 Phys. Rev. B **53**, R7642
[2] Tassone F, Piermarocchi C, Savona V, Quattropani A and Schwendimann P 1997 Phys. Rev. B **56**, 7554
[3] Tassone F and Yamamoto Y 1999 Phys. Rev. B **59**, 10830
[4] Weisbuch C, Nishioka M, Ishikawa A and Arakawa Y 1992 Phys. Rev. Lett. **69**, 3314
[5] Houdre R, Weisbuch C, Stanley R P, Oesterle U, Pellandini P and Hegems M 1994 Phys. Rev. Lett. **73**, 2043
[6] Mueller M, Bleuse J and Andre R 2000 Phys. Rev. B **62**, 16886-16892
[7] Dang L S, Heger D, Andre R, Boeuf F and Romestain R 1998 Phys. Rev. Lett. **81**, 3920
[8] Senellart P and Bloch J 1999 Phys. Rev. Lett. **82**, 1233
[9] Huang R, Tassone F and Yamamoto Y 2000 Phys. Rev. B B **61**, R7854
[10] Savvidis P G, Baumberg J J, Stevenson R M, Skolnick M S, Whittaker D M and Roberts J S 2000 Phys. Rev. Lett. **84**, 1547
[11] Stevenson R M, Astratov V N, Skolnick M S, Whittaker D M, Emam-Ismail M, Tartakovskii A I, Savvidis P G, Baumberg J J and Roberts J S 2000 Phys. Rev. Lett. **85**, 3680
[12] Senellart P, Bloch J, Sermage B and Marzin J Y 2000 Phys. Rev. B **62**, R16263
[13] Houdré R, Weisbuch C, Stanley R P, Oesterle U and Ilegems M 2000 Phys. Rev. Lett. **85**, 2793
[14] Bloch J, Sermage B, Jacquot C, Senellart P and Thierry-Mieg V 2002 phys. stat. sol. (a) **190**, 827
[15] Boeuf F, André R, Romestain R, Dang L S, Péronne E, Lampin J F, Hulin D and Alexandrou A 2000 Phys. Rev. B **62**, R22792
[16] Alexandrou A, Bianchi G, Péronne E, B. Hallé, F. Boeuf, André R, Romestain R and Dang L S 2001 Phys. Rev. B **64**, 233318
[17] Tartakovskii A I, Kulakovskii V D, Krizhanovskii D N, Skolnick M S, Astratov V N, Armitage A and Roberts J S 1999 Phys. Rev. B **60**, 11293

[18] Renucci P, Marie X, Amand T, Paillard M, Senellart P and Bloch J 2001 Springer Proceed. in Physics vol 87, ed. N. Miura and T. Ando (New York: Springer) 653.
[19] Martin M D, Viña L, Son J K and Mendez E E 2001 Solid State Commun. **117**, 267; Martín M D, Viña L. and Mendez E E 2001 Solid State Commun. **119**, 259
[20] Martín M D, Aichmayr G, Viña L and André R 2002 Phys. Rev. Lett. (in press)
[21] Aichmayr G, Martín M D, Viña L and André R 2002 (these Proceedings)
[22] Lagoudakis P G, Savvidis P G, Baumberg J J, Whittaker D M, Eastham P R, Skolnick M S and Robert J S 2002 Phys. Rev. B **65**, 161310
[23] Savvidis P G, Baumberg J J, Porras D, Whittaker D M, Skolnick S and Roberts J S 2002 Phys. Rev. B **65**, 73309
[24] Erland J, Mizeikis V, Langbein W, Jensen J R and Hvam J M 2001 Phys. Rev. Lett. **86**, 5791
[25] Dasbach G, Dremin A A, Bayer M, Kulakovskii V D, Gippius N A and Forchel A 2002 Phys. Rev. B **65**, 245316
[26] Fernández-Rossier J, Tejedor C, Muñoz L and Viña L 1996 Phys. Rev. B **54**, 11582