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Oscillatory behaviour in the nonlinear emission of semiconductor microcavities

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Abstract

We have observed marked oscillations in the time-resolved photoluminescence of a semiconductor microcavity under non-resonant excitation conditions. Hot excitons, created with an ultrashort light pulse, rapidly relax into polaritons in the cavity with a large in-plane momentum **K**. Shortly after illumination, above a certain excitation power, the polaritons accumulate into an energy trap at the bottom of the dispersion and the light emission, governed by final-state stimulated scattering, starts at $\mathbf{K} \sim 0$ (0° with respect to the normal of the sample). The angular dependence of the photoluminescence at negative detunings reveals that the emission is rapidly transferred to $\mathbf{K} \sim 2 \times 10^4$ cm^{-1} (∼15°), close to the point of inflection of the lower polariton branch, giving rise to an annular emission. The oscillations arise from a macroscopic coherent population in the lower polariton branch.

Semiconductor microcavities have been the subject of intense investigations in the last few years because they open new ways to manipulate light–matter interactions in the solid state [1]. One of the important issues to be solved is to understand the relaxation of excitons, optically pumped out of resonance ($\mathbf{K} \neq 0$, **K** being the in-plane centre of mass momentum), into cavity polaritons, its influence on a possible condensed state at $\mathbf{K} = 0$, [2] and the realization of a polariton laser [3, 4]. For that it is of crucial importance to have a precise knowledge of the emission dynamics of the polaritons at different **K** in their dispersion relation, which is completely distorted compared to that of the electron–hole pairs [5]. In this paper we present an angle-, time- and polarization-resolved photoluminescence (PL) study of a semiconductor microcavity under non-resonant excitation conditions.

The sample is a $Cd_{0.40}Mg_{0.60}Te$ microcavity of thickness λ, with top (bottom) distributed-Bragg-reflectors (DBRs), mirrors 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. The radiation–matter interaction leads to a Rabi splitting of ∼14 meV. A wedge in the cavity thickness allows tuning of the cavity and the exciton into resonance by moving the excitation spot across the wafer. The sample, kept at 2 K in an immersion cryostat, is optically excited at the first minimum above the stop-band of the DBRs (1.686 eV) with 1.5 ps pulses. The emission, angle-resolved using a small pinhole, which obtains a resolution of 1°, is detected at the lower polariton (LP) branch (1.61 eV at $\mathbf{K} = 0$) and time- and spectrally-resolved using a streak camera with an overall time resolution of ∼6 ps. For polarization-resolved measurements, a pair of $\lambda/4$ plates are included in the experiment: the excitation light is σ^+-polarized and the PL emission is analysed into its σ^+- and σ^--polarized components. The experiments are performed at negative detunings with power densities below 50 W cm^{-2}, ensuring the persistence of the strong-coupling regime.

Figure 1 depicts in a semi-logarithmic scale the time evolution of the emission at $\mathbf{K} = 0$, for a detuning of $\delta = -13$ meV, at different power densities ranging from 0.5 to 40 W cm^{-2}. The sample is excited with σ^+-polarized pulses and the emission is analysed into its co-polarized (σ^+) component. The rise-time is practically independent of the excitation power in the range used in these experiments, although a closer look at the initial curvature of the time traces reveals that this curvature changes from negative (low excitation) to positive (high excitation), as has been described

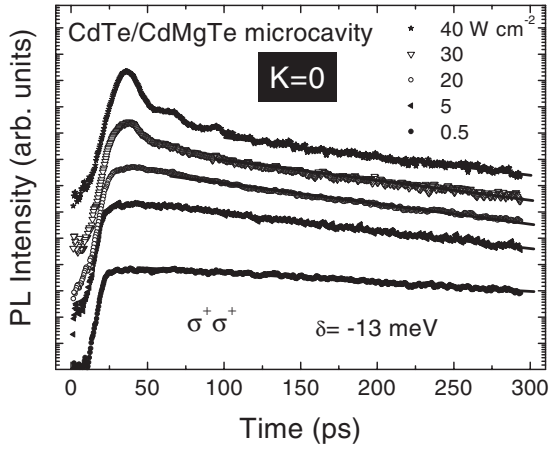

Figure 1. Time evolution, on a semi-logarithmic scale, of the PL from the LP branch at $\mathbf{K} = 0$ of the CdTe/CdMgTe microcavity for different excitation densities at a detuning of -13 meV. The excitation is σ^+-polarized and the emission is analysed into its σ^+ component.

previously [6]. A positive curvature indicates a relaxation mechanism that is based on an increasing rate, like final state stimulation [7]. The decay time decreases from ∼300 ps (at 0.5 W cm^{-2}) to ∼150 ps (at 20 W cm^{-2}) and at higher powers an additional much faster initial decay is found with a decay constant of ∼10 ps, while the slower decay remains constant at 150 ps within experimental error.

The spin dynamics and its excitation-power dependence are shown in figure 2 for emission at $\mathbf{K} = 0$. Panels (*a*) and (*c*) depict the time evolution of the emission after excitation with σ^+-polarized pulses and analysing its polarized components for 0.5 and 40 W cm^{-2}, respectively. The corresponding evolution of the degree of polarization, defined as $P = (I^{++} - I^{+-})/(I^{++} + I^{+-})$, with I^{++} (I^{+-}) being the emission intensity after σ^+ polarized excitation and σ^+ (σ^-) detection, are depicted in panels (*b*) and (*d*). At low powers, the dynamics of P is very similar to that found for bare excitons in quantum wells, except for very short times where a re-polarization occurs, which increases P from its initial value of ≈10% up to a value of 30%. This initial behaviour has also been observed in GaAs microcavities and attributed to polariton–polariton scattering [8]. At high powers, the dynamics of P is remarkably different: at very short times the degree of polarization obtains negative values and then decays towards zero, showing some oscillations in its temporal evolution. The relaxation of the non-resonantly created excitons to form polaritons, which finally emit from $\mathbf{K} = 0$, is governed by final-state stimulated scattering. Nevertheless, the scattering to the -1 spin states is more efficient than that to +1 spin states. The different scattering efficiencies might be related to an energy splitting observed between the two circularly polarized components of the PL at very short times, which is explained in detail in [9, 10].

Figure 3 shows the time evolution of the PL for different angles of detection, ranging from 0° to 15.4°, for an excitation density of 45 W cm^{-2}. Under these conditions, the strong coupling regime is still preserved. This can be inferred from the energy position of the emission from the LP branch (not shown), which at resonance ($\delta = 0$) has shifted only 2 meV towards the blue, much less than the coupling energy. Several aspects are worth discussing in the dynamical behaviour of the light emission: the strongest emission occurs at an angle of ∼15° and not at 0°; striking periodic oscillations in the emission intensity are observed, with a period independent of the observation angle; and the time delay to reach the first maximum of the emission increases monotonically with increasing angle.

The non-resonant excitation of the sample creates excitons that rapidly relax into polaritons in the cavity. The emission starts at $\mathbf{K} \sim 0$ (0°): at early times the dynamics of the light emission is governed by final-state stimulated scattering,

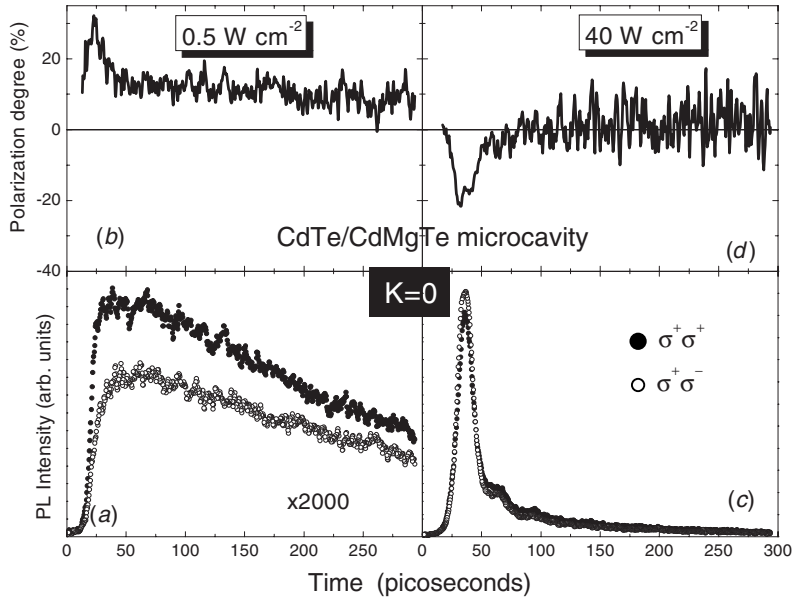

Figure 2. Time evolution, on a linear scale, of the PL from the LP branch at $\mathbf{K} = 0$ of the CdTe/CdMgTe microcavity at a detuning of -13 meV for 0.5 W cm^{-2} (*a*) and 40 W cm^{-2} (*c*); full (open) symbols correspond to co- (counter-) polarized emission to the σ^+-polarized excitation pulse. Panel (*b*) [(*d*)] depicts the time evolution of the polarization degree of the emission shown in (*a*) [(*c*)].

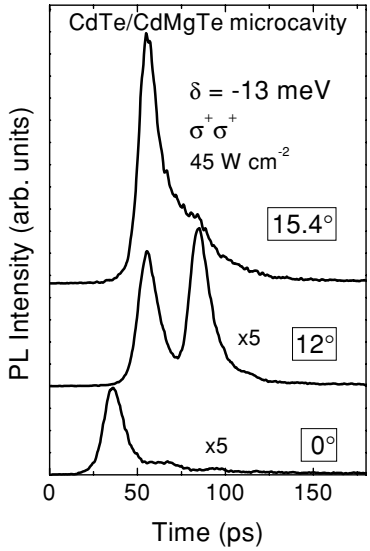

Figure 3. Time evolution traces of the PL from the LP branch at $\mathbf{K} = 0$ of the CdTe/CdMgTe microcavity at a detuning of -13 meV for different angles. The traces are enlarged by the factor shown in the figure. Note the oscillatory behaviour with a period of 30 ps.

which is responsible for the very fast rise and decay times of the PL. The bosonic character of the mechanism governing the relaxation is evidenced by an exponential increase of the PL intensity from the photon-like LP branch with excitation power, similar to that observed in pump-and-probe [11] and cw-photoluminescence experiments [12] and attributed to final-state stimulation (FSS). However, this behaviour is transient and at later times the maximum intensity of the emission occurs at an angle of 15.4° ($\mathbf{K} \sim 2 \times 10^4$ cm^{-1}), giving rise to a ring emission, which has also been observed in resonant excitation cw [13, 14] and time-resolved experiments [15]. In our case, the pulsed excitation at moderately high powers creates a large population and, therefore, the emission starts at $\mathbf{K} = 0$ assisted by FSS. The resulting decrease of population, through very fast emission dynamics, shifts the emission to larger $\mathbf{K}$ values, in agreement with the theoretical predictions [14]. More striking is the observation of strong oscillations in the emission intensity, with a period of 30 ps, which is independent of the observation angle and the helicity of the emitted light. The trace corresponding to an angle of 12° reveals that the intensity becomes stronger, for certain angles, after the first period, a behaviour never 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 136 μeV, which does not compare to any characteristic energy of our microcavity. A similar beat period, reported recently for wire-shaped microcavities [16], has been attributed to uniaxial strain in the quantum well plane resulting from the cavity patterning to form wires.

In summary, we have reported strong oscillations in the time-, polarization- and angle-resolved light emission of a CdTe microcavity when the system is optically driven into the nonlinear regime. The period of the oscillations amounts to 30 ps independent of the observation angle. The emission starts at $\mathbf{K} \sim 0$ cm^{-1} and rapidly evolves into a ring emission of radius $\mathbf{K} \sim 2 \times 10^4$ cm^{-1}.

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