

## Quantum beats between light and dark polariton states in semiconductor microcavities

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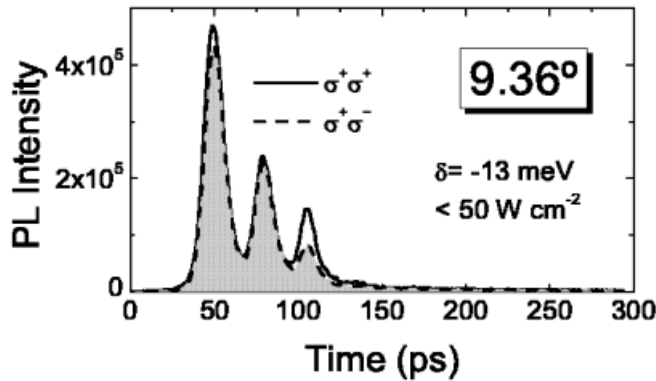
We observe experimentally pronounced beats of the intensity of photoluminescence from a bottleneck region of a microcavity in the strong coupling regime. These beats are extremely sensitive to the pumping intensity and vanish for the weak pumping. We show theoretically that the process of polariton-polariton scattering leading to the mixing between bright and dark polariton states is responsible for this effect.

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Exciton-polaritons in semiconductor microcavities have become an object of intensive experimental and theoretical studies in the recent decade. Cavity polaritons are unique excitations in solid state systems that conserve the coherence over tens of picoseconds despite both elastic and inelastic scattering events. An extremely low decoherence rate of the polaritons is because of the enhanced strength of light-matter coupling in microcavities that dominates over excitonic coupling with the phonon bath. Thanks to this effect many interesting phenomena could have been observed in the microcavities as e.g. stimulated scattering of exciton-polaritons [1] and possibly even their Bose-condensation [2]. Here we report an experimental evidence of a new quantum effect linked to the long-living optical coherence in microcavities. We have observed the quantum beats between optically active and forbidden polariton states coupled by a polariton-polariton scattering process.

Exciton-polaritons, being composite particles, retain both the properties of excitons and photons. In particular, they have the same spin-structure as excitons. In quantum wells (QWs), the lowest energy level of a heavy hole lies typically lower than any light-hole energy level, and thus the exciton ground state is formed by an electron and a heavy-hole. The entire exciton spin thus has the following projections on the structure axis:  $\pm 1$  and  $\pm 2$  allowed for the ground state. The states with the spin projection  $\pm 1$  are optically active (*bright*), as they can be excited by right or left circular polarized light beam. On the other hand, the optical excitation of  $\pm 2$  states is forbidden by the selection rules. These are so-called spin-forbidden *dark* states. Dark and bright excitonic states are typically split by a few tens of  $\mu eV$ . Mixing and coherent excitation of these states may result in oscillations of their populations, i.e. quantum beats. The mixing can be achieved by application of an in-plane magnetic field as has been experimentally demonstrated in QWs [3] and recently in microcavities [4]. Neither mixing nor beats have been observed in the absence of the field till now, to the best of our knowledge. In our microcavity system, extremely pronounced quantum beats between bright and dark states appear in the non-linear regime while *no magnetic field* is applied. On the other hand, in the linear regime we do not see any beats.

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**Fig. 1** Temporal dependence of the photoluminescence intensity from the bottleneck region for our microcavity under non-resonant strong pumping (experiment at  $9.36^\circ$ ).

Our sample is a  $\text{Cd}_{0.40}\text{Mg}_{0.60}\text{Te}$  microcavity of  $\lambda$  thickness, sandwiched between distributed Bragg reflectors (DBRs). Two pairs of  $90\text{-\AA}$  thick CdTe quantum wells are placed at the antinodes of the cavity, leading to a Rabi splitting of  $\sim 14$  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 [5].

The microcavity is mounted in an immersion cryostat 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, is time- and spectrally-resolved using a streak camera with a time resolution of  $\sim 5$  ps. For polarisation-resolved measurements, a pair of  $\lambda/4$  plates are included in the experiment: the excitation light is  $\sigma^+$ -polarised and the PL emission is analysed into its  $\sigma^+$ - and  $\sigma^-$ -polarised components. The experiments presented here are performed at a negative detuning of  $-13$  meV with power densities below  $50 \text{ W cm}^{-2}$ , which ensure that the strong-coupling regime is maintained. Only the heavy hole excitons were optically excited, the light-hole exciton resonance is not seen in the spectra as it lies at higher energies and is not coupled to the cavity mode.

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 an angle of detection of  $9.36^\circ$ . One can see that the intensity of photoluminescence experiences oscillations in both circular polarisations. Remarkably, these oscillations are *in phase*, so that they cannot be caused by any kind of beats between  $\sigma^+$  and  $\sigma^-$ -polarized polariton states. On the other hand, the oscillations we observed strongly remind the beats between bright- and dark-exciton states observed by Renucci et al [4]. In their experiment, the beats have been induced by an in-plane magnetic field which mixed exciton-polariton states with  $\pm 1$  and  $\pm 2$  spin projections on the structure axis. In our experiment, no magnetic field has been applied to the sample. On the other hand, the effect we have observed appeared to be strongly non-linear. At low pumping intensities we do not observe any beats, they only appear at strong enough pumping.

We conclude that in the non-linear regime some new mixing mechanism appears that allows for the dark states to be populated and for quantum beats between dark and bright polaritons to appear. This mechanism must conserve the spin and optical coherence in the system, otherwise quantum interference between different polariton states would be impossible.

To explain theoretically these results, let us take into account the polariton-polariton collisions described by the following interaction Hamiltonian

$$H_{\text{int}} = \begin{array}{c} \begin{array}{ccc} +2, k & & -2, k' \\ & \nearrow & \nwarrow \\ & \text{---} & \\ & \nwarrow & \nearrow \\ +1, k & & -1, k' \end{array} & + & \begin{array}{ccc} +1, k & & -1, k' \\ & \nearrow & \nwarrow \\ & \text{---} & \\ & \nwarrow & \nearrow \\ +2, k & & -2, k' \end{array} \end{array} \quad (1)$$

One can see that it conserves the spin in the system. Actually, it is an electron-electron (or hole-hole) exchange process that makes from two bright excitons with opposite spins two dark excitons with opposite spins or vice-versa.

The total Hamiltonian can be thus represented as

$$H = \sum_k \left( \varepsilon_1 p_{1\uparrow;k}^+ p_{1\uparrow;k} + \varepsilon_1 p_{1\downarrow;k}^+ p_{1\downarrow;k} + \varepsilon_2 p_{2\uparrow;k}^+ p_{2\uparrow;k} + \varepsilon_2 p_{2\downarrow;k}^+ p_{2\downarrow;k} \right) + \sum_{k,k'} W_{kk'} \left( p_{1\uparrow;k}^+ p_{1\downarrow;k'}^+ p_{2\uparrow;k} p_{2\downarrow;k'} + p_{1\uparrow;k} p_{1\downarrow;k'} p_{2\uparrow;k}^+ p_{2\downarrow;k'}^+ \right) \quad (1a)$$

where  $p_{1\uparrow\downarrow}, p_{1\uparrow\downarrow}^+$  are annihilation and creation operators for bright polaritons with opposite spin projections  $\pm 1$ ,  $p_{2\uparrow\downarrow}, p_{2\uparrow\downarrow}^+$  being the operators for dark polaritons. All the polaritons are considered being situated at the elastic circle in reciprocal space, corresponding to the bottleneck region, and thus the energies of bright states  $\varepsilon_1$  and dark states  $\varepsilon_2$  are supposed to be independent of the index  $k$ , indicating the position of a polariton at the elastic circle. The number of states  $\nu$  at the elastic circle can be estimated from the dimension of the system  $L$

$$\nu L \approx 1 \quad (2)$$

The imaginary parts was introduced into the energies, in order to take into account the finite polariton life time due to the radiative decay of bright polariton states and slow diffusion towards the ground state. The first term in Hamiltonian describes non-interacting particles; the second corresponds to the scattering of two bright polaritons with opposite spins into two dark polaritons and vice versa. Only the processes of forward scattering with electron exchange were taken into account, as the probability of hole exchange is usually much less than that for the electrons [6].

In order to obtain a set of the dynamic equations describing the temporal dependence of the polariton occupation numbers, we used the Heisenberg motion equations for the operators  $p, p^+$ :

$$i\hbar \frac{dp_{1\uparrow;l}}{dt} = [p_{1\uparrow;l}; H] = \varepsilon_1 p_{1\uparrow;l} + \sum_k W_{kl} p_{1\downarrow;k}^+ p_{2\uparrow;l} p_{2\downarrow;k} \quad (3)$$

The equations for other creation and annihilation operators are similar. Using (3), it is easy to obtain the following equations for the polariton occupation numbers  $N_{1\uparrow;l} = \langle p_{1\uparrow;l}^+ p_{1\uparrow;l} \rangle$  etc.

$$\frac{dN_{1\uparrow;l}}{dt} = -\frac{1}{\tau_{1\uparrow}} N_{1\uparrow;l} + \frac{2}{\hbar} \text{Im} \left\{ \sum_k W_{kl} \langle p_{1\uparrow;l}^+ p_{1\downarrow;k}^+ p_{2\uparrow;l} p_{2\downarrow;k} \rangle \right\} = -\frac{1}{\tau_{1\uparrow}} N_{1\uparrow;l} + \frac{2}{\hbar} \text{Im} \left\{ \sum_k W_{kl} A_{kl} \right\} \quad (4)$$

where  $\tau_1^{-1} = -2\text{Im}(\varepsilon_1)/\hbar$ . The same equations could be obtained also for  $N_{1\downarrow}, N_{2\uparrow}, N_{2\downarrow}$ . The dynamics of occupation numbers is thus determined by the dynamics of non-classical fourth order correlators  $A_{kl} = \langle p_{1\uparrow;l}^+ p_{1\downarrow;k}^+ p_{2\uparrow;l} p_{2\downarrow;k} \rangle$ , for which simple, but tiresome algebraic calculations give

$$\begin{aligned} \frac{dA_{kl}}{dt} &= \frac{2i}{\hbar} (\varepsilon_1^* - \varepsilon_2) A_{kl} + \frac{i}{\hbar} \sum_j \left\{ W_{jl} \langle p_{1\downarrow;j} p_{1\downarrow;k}^+ p_{2\uparrow;l}^+ p_{2\uparrow;l} p_{2\downarrow;j} p_{2\downarrow;k} \rangle + W_{jk} \langle p_{1\uparrow;l}^+ p_{1\uparrow;j} p_{2\uparrow;j}^+ p_{2\uparrow;l} p_{2\downarrow;k}^+ p_{2\downarrow;k} \rangle \right. \\ &\quad \left. - W_{jl} \langle p_{1\uparrow;l}^+ p_{1\uparrow;l} p_{1\downarrow;k}^+ p_{1\downarrow;j} p_{2\downarrow;j} p_{2\downarrow;k} \rangle - W_{jk} \langle p_{1\uparrow;l}^+ p_{1\uparrow;j} p_{1\downarrow;k}^+ p_{1\downarrow;k} p_{2\uparrow;l}^+ p_{2\uparrow;j} \rangle \right\} \\ &\approx \frac{2i}{\hbar} (\varepsilon_1^* - \varepsilon_2) A_{kl} + \frac{i}{\hbar} \left( W_{ll} (N_{2\uparrow;l} - N_{1\uparrow;l}) (N_{1\downarrow;k} + 1) N_{2\downarrow;k} + W_{kk} (N_{2\downarrow;k} - N_{1\downarrow;k}) N_{1\uparrow;l} (N_{2\uparrow;l} + 1) \right) \\ &\quad + \frac{i}{\hbar} \sum_{j \neq k, l} \left\{ W_{jl} (N_{2\uparrow;l} - N_{1\uparrow;l}) B_{kj;\downarrow} + W_{jk} (N_{2\downarrow;k} - N_{1\downarrow;k}) B_{kj;\uparrow} \right\} \end{aligned} \quad (5)$$

where the following approximations were used for simplifying the sixth order correlators

$$\langle p_{1\uparrow;l}^+ p_{1\uparrow;l} p_{1\downarrow;k}^+ p_{1\downarrow;k} p_{2\downarrow;j}^+ p_{2\downarrow;j} p_{2\downarrow;k} \rangle \approx N_{1\uparrow;l} \langle p_{1\downarrow;k}^+ p_{1\downarrow;k} p_{2\downarrow;j}^+ p_{2\downarrow;j} p_{2\downarrow;k} \rangle, \quad (6)$$

$$\langle p_{1\uparrow;l}^+ p_{1\uparrow;l} p_{1\downarrow;k}^+ p_{1\downarrow;k} p_{2\downarrow;j}^+ p_{2\downarrow;j} p_{2\downarrow;k} \rangle \approx N_{1\uparrow;l} N_{1\downarrow;k} N_{2\downarrow;j} \quad (6a)$$

etc.

The correlators of the new type  $B_{kj;\downarrow} = \langle p_{1\downarrow;k}^+ p_{1\downarrow;j} p_{2\downarrow;j} p_{2\downarrow;k} \rangle$ ,  $B_{kj;\uparrow} = \langle p_{1\uparrow;l}^+ p_{1\uparrow;j} p_{2\uparrow;j} p_{2\uparrow;l} \rangle$  appear in the equation for A. It is easy to show, that

$$\frac{dB_{kl;\downarrow}}{dt} = \frac{i}{\hbar} (\varepsilon_1^* - \varepsilon_1 + \varepsilon_2^* - \varepsilon_2) B_{kl;\downarrow} + \frac{i}{\hbar} \sum_j \{ W_{jk} (N_{2\downarrow;k} - N_{1\downarrow;k}) A_{lj}^* + W_{jl} (N_{1\downarrow;l} - N_{2\downarrow;l}) A_{kj} \}, \quad (7)$$

$$\frac{dB_{kl;\uparrow}}{dt} = \frac{i}{\hbar} (\varepsilon_1^* - \varepsilon_1 + \varepsilon_2^* - \varepsilon_2) B_{kl;\uparrow} + \frac{i}{\hbar} \sum_j \{ W_{jk} (N_{2\uparrow;k} - N_{1\uparrow;k}) A_{jl}^* + W_{jl} (N_{1\uparrow;l} - N_{2\uparrow;l}) A_{jk} \} \quad (8)$$

Equations (4), (5), (7), (8) represent a closed set of differential equations, which can be treated numerically. In order to understand the physical mechanisms responsible for the unusual polariton spin-dynamics in microcavities, it is instructive to consider a case when the distribution of the polaritons in the reciprocal space and the polariton-polariton scattering is isotropic, i.e. the occupation numbers, scattering coefficients and fourth order correlators do not depend on the indices k,j. The full set of the kinetic equations can be now reduced to seven equations

$$\frac{dN_{1\uparrow}}{dt} = -\frac{1}{\tau_1} N_{1\uparrow} + \frac{2}{\hbar} \text{Im}\{WA\}, \quad (9a)$$

$$\frac{dN_{1\downarrow}}{dt} = -\frac{1}{\tau_1} N_{1\downarrow} + \frac{2}{\hbar} \text{Im}\{WA\}, \quad (9b)$$

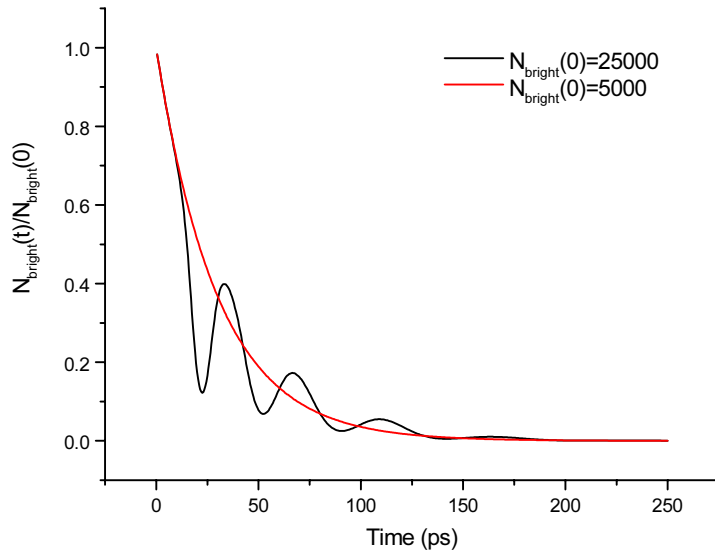
$$\frac{dN_{2\uparrow}}{dt} = -\frac{1}{\tau_2} N_{2\uparrow} - \frac{2}{\hbar} \text{Im}\{WA\}, \quad (9c)$$

$$\frac{dN_{2\downarrow}}{dt} = -\frac{1}{\tau_2} N_{2\downarrow} - \frac{2}{\hbar} \text{Im}\{WA\}, \quad (9d)$$

$$\begin{aligned} \frac{dA}{dt} = & -\left( \frac{1}{\tau_1} + \frac{1}{\tau_2} - \frac{2i}{\hbar} \Delta\varepsilon \right) A + \frac{iW}{\hbar\nu} \{ N_{2\uparrow} N_{2\downarrow} (N_{1\downarrow} + N_{1\uparrow} + \nu) - N_{1\uparrow} N_{1\downarrow} (N_{2\downarrow} + N_{2\uparrow} + \nu) \} \\ & + \frac{iW}{\hbar} \{ (N_{2\uparrow} - N_{1\uparrow}) B_{\downarrow} + (N_{2\downarrow} - N_{1\downarrow}) B_{\uparrow} \}, \end{aligned} \quad (9e)$$

$$\frac{dB_{\downarrow}}{dt} = -\left( \frac{1}{\tau_1} + \frac{1}{\tau_2} \right) B_{\downarrow} + \frac{2iW}{\hbar} (N_{2\downarrow} - N_{1\downarrow}) \text{Im}\{A\}, \quad (9f)$$

$$\frac{dB_{\uparrow}}{dt} = -\left( \frac{1}{\tau_1} + \frac{1}{\tau_2} \right) B_{\uparrow} + \frac{2iW}{\hbar} (N_{2\uparrow} - N_{1\uparrow}) \text{Im}\{A\} \quad (9g)$$



**Fig. 2** Results of the numerical solution of Eqs. (9) for strong and weak pumping at the observation angle of  $9.36^\circ$ .

where  $N = \sum_{k=1}^{\nu} N_k$ ,  $A = \sum_{k,l=1}^{\nu} A_{kl}$ ,  $B = \sum_{k,l=1}^{\nu} B_{kl}$ ,  $\Delta\varepsilon = \text{Re}(\varepsilon_1 - \varepsilon_2)$ ,  $\nu$  is a number of states at the elastic circle corresponding to bottleneck region. The matrix element of the interaction can be roughly estimated as [7]

$$W \sim E_B a_B^2 / S \quad (10)$$

where  $E_B$  is the exciton binding energy,  $a_B$  is an exciton radius,  $S$  is an area of the sample.

For our numerical calculations we have chosen the following values of the parameters:  $W = 1.2 \mu\text{eV}$ , which corresponds to the diameter of the laser spot of tenths of micrometers,  $\Delta\varepsilon = 0.04 \text{ meV}$ ,  $\nu = 1000$ ,  $\tau_1 = 30 \text{ ps}$ ,  $\tau_2 = \infty$ . All the polaritons were supposed to occupy equally two bright states at the beginning. The initial absence of the dark excitons in the system is a natural assumption, as they can not be created by the exciting light beam. Putting the occupation numbers of the two bright states equal we assumed that the spin relaxation in the system is fast enough to equalize them during the relaxation along the exciton-like branch [8].

Figure 2 shows the result of the numerical solution of the Eqs. (9) for two initial values of the initial number of polaritons  $N_0$  determined by the pump intensity. One can see that for high values of  $N_0$  pronounced oscillations of the occupation numbers of bright states, and, consequently, of the photoluminescence intensity, appear. The period of these oscillations corresponds well to the experimentally observed. On the other hand, for the weaker pump intensity no oscillations appear and one observes a monotonous decay of the photoluminescence intensity governed by the processes of radiative decay and slow scattering of the polaritons towards the ground state.

In conclusion, we have discovered experimentally and described theoretically a new mechanism of quantum oscillations in a semiconductor microcavity. The beats between spin-allowed (“bright”) and spin-forbidden (“dark”) exciton-polariton states take place in a non-linear regime due to polariton-polariton collisions. Remarkably, these collisions conserve the spin and coherence in the system that allows for observation of the quantum beats.

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