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# Non-linear coupling of polariton and dark exciton states in semiconductor microcavities

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**Abstract**

We report a new mechanism of non-linear coupling of optically active and dark crystal states. We observe experimentally pronounced beats of the intensity of photoluminescence from a bottleneck region of the exciton-polariton band in a microcavity in the strong coupling regime and at strong pumping. These beats are extremely sensitive to the pumping intensity and vanish for weak pumping. We show theoretically that coherent polariton–polariton scattering which leads to the mixing between bright and dark exciton states can be responsible for this effect.


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Since the first observation of the strong coupling regime [1], semiconductor microcavities have become an object of intensive experimental and theoretical studies. Due to the strong coupling between the excitonic and photonic modes, the eigenstates of the microcavities are neither excitons nor photons, but the combination of both, known as exciton–polaritons [2]. Enhanced strength of light–matter interaction in microcavities that dominates over excitonic coupling with the phonon bath results in the extremely low decoherence rate of the polaritons, which conserve their coherence over tens of picoseconds despite both elastic and inelastic scattering events [3]. Thanks to this effect many interesting phenomena have been observed in the microcavities as e.g. stimulated scattering of exciton–polaritons [4] and possibly even their Bose-condensation [5].

Recent experimental studies have revealed strong optical nonlinearities in quantum microcavities that originate from polariton–polariton interactions. As an example, parametric amplification of polariton–polariton scattering in case of the resonant optical pumping of the low-polariton dispersion branch has been observed [4] and theoretically described [6,7]. Here we present experimental evidence of a new non-linear effect linked to polariton–polariton interactions. We have observed quantum beats between optically active polariton states and spin-forbidden exciton states coupled by a polariton–polariton (exciton–exciton) scattering process. This observation is a manifestation of a new kind of parametric oscillation process in microcavities that does not require resonant pumping and keeps coherence of the polariton (exciton) system much longer than the polariton radiative life-time (up to 100 ps in our experiment). It shows

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the remarkable potentiality of microcavities for realisation of quantum optical devices.

As the energy of a heavy-hole is typically lower than that of a light-hole, in most quantum wells (QWs) the exciton ground state is formed by an electron (spin $S=\pm 1/2$) and a heavy-hole (spin $J=\pm 3/2$. The entire exciton spin thus has $S+J=\pm 1$ and $S+J=\pm 2$ projections on the structure axis 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. They are coupled with a photonic mode of the cavity and participate in formation of the exciton–polaritons. On the other hand, the optical excitation of $\pm 2$ states is forbidden by the selection rules. These are so-called dark states, completely decoupled from the photonic modes. Dark and bright exciton states are typically split by a few tens of μeV. 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 [8] and recently in microcavities [9]. This is a linear optical effect which does not involve any parametric oscillation. In the following lines we describe it theoretically in order to reveal the difference from our own experiment which will be presented afterwards. A model Hamiltonian, describing the ensemble of cavity polaritons and dark excitons subjected to an external in-plane magnetic field writes:

$$\begin{aligned} H &= \varepsilon_1(p_{1\uparrow}^{+}p_{1\uparrow} + p_{1\downarrow}^{+}p_{1\downarrow}) + \varepsilon_2(p_{2\uparrow}^{+}p_{2\uparrow} + p_{2\downarrow}^{+}p_{2\downarrow}) \\ &+ g_e\mu_B(Bp_{2\uparrow}^{+}p_{1\uparrow} + B^{*}p_{2\downarrow}^{+}p_{1\downarrow} + B^{*}p_{1\uparrow}^{+}p_{2\uparrow} \\ &+ Bp_{1\downarrow}^{+}p_{2\downarrow}) + g_h\mu_B(Bp_{2\uparrow}^{+}p_{1\downarrow} + B^{*}p_{2\downarrow}^{+}p_{1\uparrow} \\ &+ B^{*}p_{1\downarrow}^{+}p_{2\uparrow} + Bp_{1\uparrow}^{+}p_{2\downarrow}) \end{aligned} \tag{1}$$

where $B=B_x-\mathrm{i}B_y$, $g_e$ and $g_h$ are electron and hole $g$-factors respectively, $p_{1\uparrow\downarrow}, p_{1\uparrow\downarrow}^{+}$ are annihilation and creation operators for bright polaritons with spin projections $\pm 1$, $p_{2\uparrow\downarrow}$, $p_{2\uparrow\downarrow}^{+}$ being the creation and annihilation operators for dark excitons with different spin projections $\pm 2$. The energies $\varepsilon_1$ and $\varepsilon_2$ are complex, their imaginary parts are inversely proportional to the polariton lifetimes. Using the Heisenberg equation of motion for the creation and annihilation operators $\mathrm{d}p_{i,\uparrow\downarrow}/\mathrm{d}t=\mathrm{i}\hbar^{-1}[H,p_{i,\uparrow\downarrow}]$, it is easy to obtain the following system of the kinetic equations for occupation numbers

$$\begin{aligned} \frac{\mathrm{d}N_{1\uparrow}}{\mathrm{d}t} &= -\frac{1}{\tau_1}N_{1\uparrow} \\ &+ \frac{2}{\hbar}\mathrm{Im}\left[g_e\mu_B B^{*}\langle p_{1\uparrow}^{+}p_{2\uparrow}\rangle + g_h\mu_B B\langle p_{1\uparrow}^{+}p_{2\downarrow}\rangle\right] \end{aligned} \tag{2}$$

where the polariton lifetimes $\tau_i^{-1}=-2\mathrm{Im}(\varepsilon_i)/\hbar$ are introduced. Here $N_{1\uparrow}$ is the population of the bright state with the spin projection $+1$. The equations for $N_{1\downarrow}$, $N_{2\uparrow}$, $N_{2\downarrow}$ (populations of the states with spin projections $-1$, $+2$, and $-2$, respectively) can be obtained by changing the indices in (2). The dynamics of occupation numbers is thus determined by the dynamics of non-classical second order correlators $\langle p_{1\uparrow}^{+}p_{2\uparrow}\rangle, \langle p_{1\uparrow}^{+}p_{2\downarrow}\rangle, \langle p_{1\downarrow}^{+}p_{2\uparrow}\rangle, \langle p_{1\downarrow}^{+}p_{2\downarrow}\rangle$ for which simple algebraic calculations give

$$\begin{aligned} \frac{\mathrm{d}}{\mathrm{d}t}\langle p_{1\uparrow}^{+}p_{2\uparrow}\rangle &= \frac{i}{\hbar}(\varepsilon_1^{*} - \varepsilon_2)\langle p_{1\uparrow}^{+}p_{2\uparrow}\rangle \\ &+ \frac{\mathrm{i}\mu_B}{\hbar}\left[g_eB(N_{2\uparrow} - N_{1\uparrow}) + g_h\left(B^{*}\langle p_{2\uparrow}^{+}p_{2\downarrow}\rangle^{*} - B\langle p_{1\uparrow}^{+}p_{1\downarrow}\rangle\right)\right] \end{aligned} \tag{3}$$

and similar expressions for $\langle p_{1\uparrow}^{+}p_{2\downarrow}\rangle, \langle p_{1\downarrow}^{+}p_{2\uparrow}\rangle, \langle p_{1\downarrow}^{+}p_{2\downarrow}\rangle$.

The system (2–3) is equivalent to the equations for four coupled damped harmonic oscillators and readily describes the beats between exciton–polariton states with spin $\pm 1$ and dark excitons states with spin $\pm 2$ induced by an in-plane magnetic field. In the experiment, these beats manifest themselves in the oscillations of the intensity of photoluminescence. The period of the oscillations is determined by the magnetic field only, and does not depend on the exciton–polariton concentration, i.e. the observed effect is linear [8,9].

To the best of our knowledge, neither mixing nor beats have been observed in the absence of a magnetic field till now. However, in our microcavity system, at large negative detuning, extremely pronounced oscillations appear in the non-linear regime while no magnetic field is applied. On the other hand, in the linear regime these oscillations vanish. We theoretically describe these oscillations considering quantum beats between bright and dark states as a parametric process governed by the polariton–polariton (exciton–exciton) scattering.

Our sample is a $Cd_{0.40}Mg_{0.60}Te$ microcavity of thickness $\lambda$, sandwiched between distributed Bragg reflectors (DBRs). Two pairs of 90-Å 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 [10].

The microcavity is mounted in an immersion cryostat at 1.2 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, is time- and spectrally-resolved using a streak camera with an overall time resolution of $\sim$10 ps. For polarisation-resolved measurements, two $\lambda/4$ plates are included in the experiment: the excitation light is $\sigma^{+}$-polarised and the PL emission is analysed into its $\sigma^{+}$- and $\sigma^{-}$-polarized components. The experiments presented here are performed at a negative detuning of $-13$ meV with power densities below 50 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.

Fig. 1 shows the time-evolution of the photoluminescence, excited at the first minimum above the stop-band (1.686 eV) and detected at the lower polariton branch (1.61 eV), for an angle of detection of 9.4°. Note, that all the curves in Fig. 1 are drawn in a linear scale. At low pumping intensity, one can see a monotonous decay—solid line in Fig. 1(a). With increasing power an oscillation is clearly observed in the temporal trace, which presents two maxima at 32 and 60 ps—dash-dotted line in Fig. 1(a). At higher pumping intensity (Fig. 1(b)), the PL intensity exhibits 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. The intensities of $\sigma^+$ and $\sigma^-$—components of the PL signal are rather similar, indicating that spin-relaxation of polaritons was quite efficient and that their $\pm 1$ spin states are nearly equally populated. However, the population of these two 'bright' states oscillate with a period of about 30 ps. These oscillations survive over 100 ps, which exceeds by two orders of magnitude the radiative life-time of exciton–polaritons in the photon-like part of their lower dispersion branch. The long duration of the oscillations is due to the strong bottleneck effect for large negative detunings, as we discuss later.

These oscillations closely remind the beats between bright- and dark-exciton states observed by Renucci et al. [9]. But in contrast with the Renucci experiment, no magnetic field has been applied to the sample in our case.

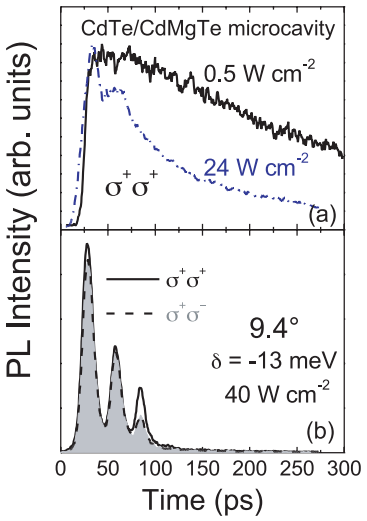


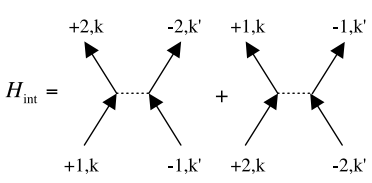

Fig. 1. Measured temporal dependence of the photoluminescence intensity from the bottleneck region (at 9.4°) of our microcavity under non-resonant pumping. (a) Solid line: low-excitation (0.5 W $cm^{-2}$); dot-dashed line: intermediate-excitation (24 W $cm^{-2}$). (b) High-excitation (40 W $cm^{-2}$): the solid and dashed lines correspond to $\sigma^+$ and $\sigma^-$—polarized emission, respectively. The pumping light is always $\sigma^+$-polarized.

Also, the effect we have observed is strongly non-linear: at low pumping intensities we do not observe any beats, they only appear at strong enough pumping.

These experimental facts make us conclude that in the non-linear regime some new mixing mechanism appears that allows for the dark states to be populated and leads to quantum beats between dark excitons and exciton–polaritons. This mechanism must conserve the spin and optical coherence in the system; otherwise quantum interference between different states would be impossible. We believe that a possible mechanism can be exchange interaction between polaritons or dark excitons with opposite spins. Let us assume that a couple of polaritons with spins $+1$ ($S=-1/2$, $J=3/2$) and $-1$ ($S=1/2$, $J=-3/2$) exchange their electrons. The resulting states will have spins $+2$ ($S=1/2$, $J=3/2$) and $-2$ ($S=-1/2$, $J=-3/2$), respectively. Thus a couple of dark excitons appears. A new exchange of electrons makes from them a couple of bright polaritons again, etc. The hole exchange would yield the same result, but as it is much less probable [11], we shall neglect it in the following discussion.

The energies of polaritons and dark excitons are split by about 0.04 meV. This splitting between dark and bright states scales as the exciton binding energy, that is why it is approximately 2.5 times larger in our CdTe based structure than in the GaAs based cavity from Ref. [8]. The coupling between dark and bright exciton states comes from the exciton–exciton scattering, therefore, it is strongly sensitive to the concentration of excitons in the given quantum state. Following Tassone [12], we estimate the matrix element of the exciton–exciton scattering as $W \tilde{} E_B a_B^2/S$; where $E_B$ is the exciton binding energy, $a_B$ is an exciton radius, $S$ is the area of the system that is assumed to be given by the spot size of the incident laser, which yield $W \approx 1.2$ μeV in our case. Achieving populations of about 30 excitons (polaritons) per quantum state renders the coupling strength comparable with the splitting between dark and bright states.

Our experiment has been performed under non-resonant pumping. The exciton-like part of the lower-polariton branch is first populated. Then, the polaritons relax toward the bottleneck region of the dispersion where they can dwell a few tens of picoseconds before proceeding towards the photon-like part of the dispersion from where they escape radiatively (see inset in Fig. 2). The beats between bright and dark states above the bottleneck lead to oscillations of the bright polariton population at the bottleneck and, consequently, in all lower states. That is why we observe oscillations of the PL intensity having the same phase for its $\sigma^+$ and $\sigma^-$ components. To describe the dynamics of such a process we introduce the following interaction Hamiltonian




(4)

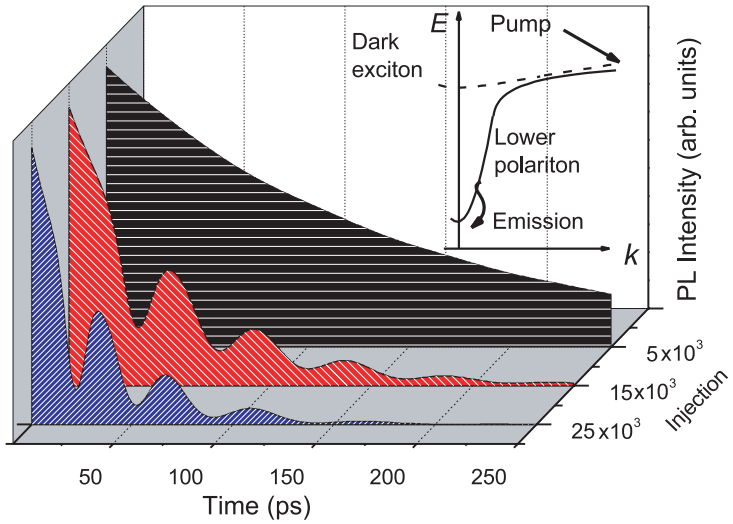



Fig. 2. Calculated intensity of the photoluminescence at 9.4° from the microcavity under study in the strong pumping ($25\times10^3$ injected polaritons,), intermediate pumping ($15\times10^3$ injected polaritons) and weak pumping ($5\times10^3$ injected polaritons) regimes. The inset shows schematically the dark-exciton and low-polariton branches.

The diagrams in the right part of Eq. (4) represent transformations of two polaritons into two dark excitons and vice-versa.

Using the second quantization technique, the total model Hamiltonian accounting for this coupling between polariton and dark exciton states can be written as:

$$H = \sum_k (\varepsilon_1 p^+_{1\uparrow;k} p_{1\uparrow;k} + \varepsilon_1 p^+_{1\downarrow;k} p_{1\downarrow;k} + \varepsilon_2 p^+_{2\uparrow;k} p_{2\uparrow;k} + \varepsilon_2 p^+_{2\downarrow;k} p_{2\downarrow;k}) + \sum_{k,k'} W_{kk'} (p^+_{1\uparrow;k} p^+_{1\downarrow;k'} p_{2\uparrow;k} p_{2\downarrow;k'} + p_{1\uparrow;k} p_{1\downarrow;k'} p^+_{2\uparrow;k} p^+_{2\downarrow;k'}) \quad (5)$$

Here we consider only the polaritons situated at the elastic circle in reciprocal space that corresponds to the exciton-like part of the dispersion branch above the bottleneck. The energies of bright states $\varepsilon_1$ and dark states $\varepsilon_2$ are supposed to be independent of the index $k$, which is related to the position of the particle on the elastic circle. $W_{kk'}$ is the matrix element of interaction, which is assumed to be given by the exciton–exciton matrix element of interaction. An imaginary part is introduced into the energies in order to take into account the finite polariton lifetime due to the radiative decay of polariton states and their energy relaxation towards lower energy states. The energy and momentum relaxation dynamics of the polaritons is accounted for in this life-time only, which is a simplifying assumption, of course.

The dynamics of occupation numbers of dark exciton and polariton states can be obtained using the Heisenberg equation. They read:

$$\frac{dN_{1\uparrow;l}}{dt} = -\frac{1}{\tau_{1\uparrow}} N_{1\uparrow;l} + \frac{2}{\hbar} \mathrm{Im} \left\{ \sum_k W_{kl} \langle p^+_{1\uparrow;l} p^+_{1\downarrow;k} p_{2\uparrow;l} p_{2\downarrow;k} \rangle \right\} \quad (6)$$

Similar equations write for $N_{1\downarrow}$,$N_{2\uparrow}$,$N_{2\downarrow}$. The dynamics of occupation numbers is governed by the dynamics of non-classical fourth order correlators $A_{kl} \equiv \langle p^+_{1\uparrow;l} p^+_{1\downarrow;k} p_{2\uparrow;l} p_{2\downarrow;k} \rangle$, for which simple, but tiresome algebraic calculations give

$$\begin{aligned}
\frac{dA_{kl}}{dt} &= \frac{2i}{\hbar}(\varepsilon_1^* - \varepsilon_2) A_{kl} + \frac{i}{\hbar} \times \sum_j \{ W_{jl} \langle p_{1\downarrow;j} p^+_{1\downarrow;k} p^+_{2\uparrow;l} p_{2\uparrow;l} p^+_{2\downarrow;j} p_{2\downarrow;k} \rangle \\
&+ W_{jk} \langle p^+_{1\uparrow;l} p_{1\uparrow;j} p^+_{2\uparrow;j} p_{2\uparrow;l} p^+_{2\downarrow;k} p_{2\downarrow;k} \rangle \\
&- W_{jl} \langle p^+_{1\uparrow;l} p_{1\uparrow;l} p^+_{1\downarrow;k} p_{1\downarrow;j} p^+_{2\downarrow;j} p_{2\downarrow;k} \rangle \\
&- W_{jk} \langle p^+_{1\uparrow;l} p_{1\uparrow;j} p^+_{1\downarrow;k} p_{1\downarrow;k} p_{2\uparrow;l} p^+_{2\uparrow;j} \rangle \} \\
&\approx \frac{2i}{\hbar}(\varepsilon_1^* - \varepsilon_2) A_{kl} + \frac{i}{\hbar} (W_{ll}(N_{2\uparrow;l} - N_{1\uparrow;l}) \\
&\times (N_{1\downarrow;k} + 1) N_{2\downarrow;k} + W_{kk}(N_{2\downarrow;k} - N_{1\downarrow;k}) N_{1\uparrow;l} \\
&\times (N_{2\uparrow;l} + 1)) + \frac{i}{\hbar} \sum_{j \neq k,l} \{ W_{jl}(N_{2\uparrow;l} - N_{1\uparrow;l}) B_{kj;\downarrow} \\
&+ W_{jk}(N_{2\downarrow;k} - N_{1\downarrow;k}) B_{lj;\uparrow} \} \quad (7)
\end{aligned}$$

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

$$\langle p^+_{1\uparrow;l} p_{1\uparrow;l} p^+_{1\downarrow;k} p_{1\downarrow;j} p^+_{2\downarrow;j} p_{2\downarrow;k} \rangle \approx N_{1\uparrow;l} \langle p^+_{1\downarrow;k} p_{1\downarrow;j} p^+_{2\downarrow;j} p_{2\downarrow;k} \rangle \quad (8)$$

$$\langle p^{+}_{1\uparrow;l} p_{1\uparrow;l} p^{+}_{1\downarrow;k} p_{1\downarrow;k} p^{+}_{2\downarrow;k} p_{2\downarrow;k} \rangle \approx N_{1\uparrow;l} N_{1\downarrow;k} N_{2\downarrow;k} \tag{8a}$$

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 $A_{kl}$. Their dynamics is given by

$$\frac{dB_{kl;\uparrow,\downarrow}}{dt} = \frac{i}{\hbar}(\varepsilon_1^* - \varepsilon_1 + \varepsilon_2^* - \varepsilon_2) B_{kl;\uparrow,\downarrow} + \frac{i}{\hbar} \times \sum_{j \neq l,k} \{ W_{jk}(N_{2\uparrow,2\downarrow;k} - N_{1\uparrow,1\downarrow;k}) A^*_{jl} + W_{jl}(N_{1\uparrow,1\downarrow;l} - N_{2\uparrow,2\downarrow;l}) A_{jk} \} \tag{9}$$

Eqs. (6), (7), and (9), represent a closed set of differential equations, which can be treated numerically. If the distribution of the polaritons in the reciprocal space and the polariton–polariton scattering are isotropic, which is a good approximation in the case of non-resonant pumping, 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 the system

$$\frac{dN_{1,2,\uparrow,\downarrow}}{dt} = -\frac{1}{\tau_{1,2}} N_{1,2,\uparrow,\downarrow} + \frac{2}{\hbar} \mathrm{Im}\{WA\} \tag{10}$$

$$\frac{dA}{dt} = -\left(\frac{1}{\tau_1} + \frac{1}{\tau_2} - \frac{2i}{\hbar}\Delta\varepsilon\right) A + \frac{iW}{\hbar\nu} \{ N_{2\uparrow} N_{2\downarrow} (N_{1\downarrow} + N_{1\uparrow} + \nu) - N_{1\uparrow} N_{1\downarrow} (N_{2\downarrow} + N_{2\uparrow} + \nu) \} + \frac{iW}{\hbar} \times \{ (N_{2\uparrow} - N_{1\uparrow}) B_{\downarrow} + (N_{2\downarrow} - N_{1\downarrow}) B_{\uparrow} \} \tag{10a}$$

$$\frac{dB_{\uparrow,\downarrow}}{dt} = -\left(\frac{1}{\tau_1} + \frac{1}{\tau_2}\right) B_{\uparrow,\downarrow} + \frac{2}{\hbar} (N_{2\uparrow,2\downarrow} - N_{1\uparrow,1\downarrow}) \mathrm{Im}\{WA\} \tag{10b}$$

where $N = \sum_{k=1}^{\nu} N_k$, $A = \sum_{k,l=1}^{\nu} A_{kl}$, $B = \sum_{k,l=1}^{\nu} B_{kl}$, $\Delta\varepsilon = \mathrm{Re}(\varepsilon_1 - \varepsilon_2)$, $\nu$ is the number of states on the elastic circle corresponding to the bottleneck region.

In the numerical calculations we have chosen $\Delta\varepsilon = 0.04$ meV, $\nu = 1000$, $\tau_1 = 150$ ps (50 ps, 30 ps) for the weak (intermediate, strong) pumping and $\tau_2 = \infty$. For simplicity, all the polaritons were supposed to occupy equally $\sigma^+$ and $\sigma^-$—polarized bright states at $t = 0$ (in the experiment, an initial polarization degree of about 20% was present at low pumping, which is not essential for the effect we describe). The concentration of dark excitons is zero at $t = 0$ as they are not created by the excitation beam.

Fig. 2 shows the calculated time evolution of the photoluminescence in the weak- and strong-pumping regimes. The traces result from the numerical solution of Eqs. (10)–(10b). The strong- and weak-pumping regimes correspond to different concentrations of polaritons $N_0$ injected to the system at $t = 0$. We took $N_0 = 25 \times 10^3$ for the strong pumping, $N_0 = 15 \times 10^3$ for the intermediate pumping and $N_0 = 5 \times 10^3$ for the weak pumping limit, respectively. One can see that in the first case pronounced oscillations of the occupation numbers of bright states, and, consequently, of the photoluminescence intensity, appear. The initial period of these oscillations is close to the experimentally observed of $\sim 30$ ps. The oscillations are still seen at the intermediate pumping, while in the weak pumping limit, no oscillations appear and a monotonous decay of the photoluminescence intensity is obtained, which is governed by the processes of radiative decay and slow scattering of the polaritons towards the ground state. These features are in qualitative agreement with our experimental observations. The model we use is the simplest one, allowing describing quasi-analytically a new effect we have observed. A detailed agreement can be achieved if one takes into account all the complicated dynamics of the polariton relaxation in microcavities solving the complete set of kinetic equations [13] and taking into account the dark exciton states. It should be mentioned that we observe the beats at different angles, and different points on the sample, although they are very sensitive to the detuning and therefore they vanish rapidly when the detuning is varied; in fact this detuning dependence supports our interpretation because the coupling with the dark states is changed.

In conclusion, we have revealed experimentally quantum oscillations in a semiconductor microcavity and provided a possible theoretical description of the responsible mechanism leading to the oscillations. The beats between exciton–polaritons and spin-forbidden ('dark') exciton states take place in a non-linear regime due to polariton–polariton and exciton–exciton collisions. Remarkably, these collisions conserve the spin and coherence in the system that allows for observation of a kind of optical parametric oscillations. The long-living coherence in our system is a consequence of the strong bottleneck effect that makes polaritons to spend the most part of their life-time in the exciton-like part of their lower dispersion branch having a low probability of radiative escape.

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