Quantum Theory of Spin Dynamics of Exciton-Polaritons in Microcavities

K.V. Kavokin,[1] I. A. Shelykh,[2,*] A.V. Kavokin,[2] G. Malpuech,[2] and P. Bigenwald[2]

[1]*A. F. Ioffe Physico-Technical Institute, 26, Politechnicheskaya, 194021 St. Petersburg, Russia*

[2]*LASMEA, UMR 6602 CNRS, Université Blaise Pascal, 24, avenue des Landais, 63177 Aubière, France*

(Received 25 July 2003; published 6 January 2004)

We present the quantum theory of momentum and spin relaxation of exciton-polaritons in microcavities. We show that giant longitudinal-transverse splitting of the polaritons mixes their spin states, which results in beats between right- and left-circularly polarized photoluminescence of microcavities, as was recently experimentally observed [M. D. Martin *et al.*, Phys. Rev. Lett. **89**, 077402 (2002)]. This effect is strongly sensitive to the bosonic stimulation of polariton scattering.

DOI: 10.1103/PhysRevLett.92.017401 PACS numbers: 78.67.–n, 42.55.Sa, 71.36.+c, 72.25.Rb

Semiconductor microcavities now attract enhanced attention due to their potential applications in optoelectronic devices of the new generation [1]. Bosonic stimulation of scattering of exciton-polaritons in microcavities paves the way to the realization of polariton lasers [2]. Exciton-polaritons are known to keep their coherence while relaxing in energy and momentum, which shows a high potentiality of microcavities for applications as optical and quantum memory elements [3]. Recent experiments have revealed a very peculiar spin dynamics of polaritons in microcavities [4–6] while no adequate theory accounting for both bosonic properties of polaritons and their spin has been reported until now, to the best of our knowledge. In this Letter, we present a model describing energy and spin relaxation of exciton-polaritons in realistic microcavities. It allows one to understand the unusual polarization beats recently observed in the emission of microcavities [6].

Among the principal mechanisms of spin relaxation of charge carriers in semiconductors, the most important are the Elliott-Yafet [7], Bir-Pikus [8], and Dyakonov-Perel [9] mechanisms connected with the intrinsic symmetry properties of crystals, spin-orbit and exchange interactions. The exciton spin in quantum wells (QWs) has ± 1 and ± 2 projections on the structure axis allowed for the ground state. $\sigma^+$and σ^- circularly polarized light excites $+1$ and -1 excitons, respectively. Linearly polarized light excites a linear combination of $+1$ and -1 exciton states, so that the total exciton-spin projection on the structure axis is zero in this case. Because of the splitting between ± 1 and ± 2 doublets, the spin-relaxation mechanisms listed above are less effective for excitons and, more importantly, new strong mechanisms arise. The most universal of them, referred to as the Maialle mechanism (Ref. [10]), is due to the longitudinal-transverse splitting of the radiative exciton doublet. We discuss this mechanism below for the case of exciton-polaritons, in which it is expected to be especially strong. Also, the exciton spin relaxation may be realized via spin flips due to collisions of excitons with structure imperfections, impurities, or other excitons.

Exciton-polaritons have the same spin structure as excitons. The difference between their spin-relaxation dynamics and spin relaxation of bare excitons was expected to come from the different shape of dispersion curves and, consequently, different energy relaxation dynamics.

Exciton spin dynamics in semiconductors can be experimentally studied by measurement of time-resolved polarization of light emitted by excitons. The polarization degree of light $\wp$ is given by the difference between the concentrations of spin $+1$ and -1 excitons (N^+ and N^-, respectively) normalized to the total exciton concentration:

$$\wp = \frac{N^+ - N^-}{N^+ + N^-}. \tag{1}$$

The goal of this work is to calculate the polarization degree of the photoluminescence from polariton states in a microcavity in the strong coupling regime.

It is convenient to treat polariton relaxation in microcavities using the pseudospin formalism [11]. The pseudospin vector $\vec{S}$ is defined on the two quantum states belonging to the radiative exciton (polariton) doublet. It describes both the exciton-spin state and its dipole moment orientation. The density matrix of the system can be expressed as [11]

$$\rho_{\vec{k}} = \frac{N_{\vec{k}}}{2} I + \vec{S}_{\vec{k}} \cdot \vec{\sigma}, \tag{2}$$

where I is the identity matrix, $\vec{\sigma}$ is the Pauli-matrix vector, and $\vec{S}_{\vec{k}}$ is the mean pseudospin of the polaritons with the wave vector $\vec{k}$. It corresponds to the Poincaré vector of partially polarized light (see Fig. 1). The general form of the polariton Hamiltonian in terms of pseudospin reads

$$H_{\vec{k}} = E_n(\vec{k}) + \vec{\Omega}_n(\vec{k}) \cdot \vec{S}_{\vec{k}}, \tag{3}$$

where $E_n(\vec{k})$ is the energy of the nth polariton branch, and $\vec{\Omega}_n(\vec{k})$ is an effective magnetic field. The z component of $\vec{\Omega}_n(\vec{k})$, which splits $+1$ and -1 exciton states, is the real

 © 2004 The American Physical Society

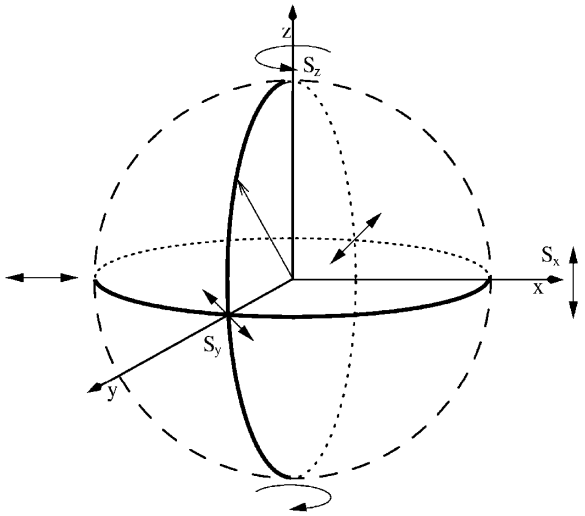

FIG. 1. A Poincaré sphere with a pseudospin. The equator of the sphere corresponds to different linear polarizations, while the poles correspond to two circular polarizations.

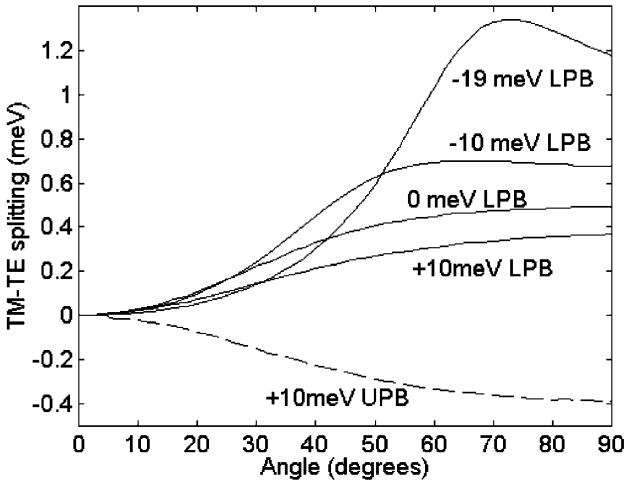

FIG. 2. Longitudinal-transverse (TM-TE) polariton splitting calculated for a microcavity sample from Ref. [6] for the lower polariton branch (LPB) and the upper polariton branch (UPB) for different detunings.

magnetic field normal to the structure plane. X and Y components of $\vec{\Omega}_n(\vec{k})$ are nonzero if the exciton states having dipole moments in, say, x and y directions have different energies. This is typically the case for excitons having nonzero in-plane wave vectors. The splitting of exciton states with dipole moments parallel and perpendicular to the wave vector is called longitudinal-transverse splitting. It results from the long-range exchange interaction. The splitting is zero for $\vec{k} = 0$ and increases as a function of k, following a square root law at large k [12].

In microcavities, the splitting of longitudinal and transverse polariton states is amplified due to the exciton coupling with a cavity mode. Note that the cavity mode frequency is also split in TE- and TM-light polarizations [13]. The resulting polariton splitting strongly depends on the detuning between the cavity mode and the exciton resonance and, in general, increases with k. Figure 2 shows the longitudinal-transverse polariton splitting Ω_k calculated for a microcavity sample from Ref. [6] at different detunings. Polariton eigenfrequencies in the two polarizations have been found numerically by a transfer matrix method. One can see that the splitting is very sensitive to the detuning and has different signs for the upper and lower polariton branches. The difference in signs comes from different exciton oscillator strengths causing different Rabi splittings in TE and TM polarizations.

We now proceed with deriving the kinetic equation for polarized polariton ensembles. The populations of polaritons having pseudospin projections $\pm 1/2$ onto the axis of the structure $N_{\vec{k}}^{\pm}$ are given by the diagonal elements of the density matrix (2). Consider scattering of polaritons from a quantum state 1 to the quantum state 2. As the spin is conserved during the scattering act, the variation of population of polaritons in state 2 can be written as

$$\left(\frac{dN_2^{\pm}}{dt}\right)_{1\to 2} = W_{12} N_1^{\pm}(N_2^{\pm} + 1), \tag{4}$$

where W_{12} describes the scattering probability from quantum state 1 to quantum state 2.

Equation (4) describes the variation of the diagonal elements of the density matrix (2) if all pseudospins are parallel to the z axis. The dynamics of the off-diagonal elements of the density matrix is less trivial. To obtain it within the same approximations as above, let us consider the evolution of the density matrix ρ_2, describing state 2, due to the scattering from state 1 as a result of the linear transformation of ρ_1:

$$\left(\frac{\partial \rho_2}{\partial t}\right)_{1\to 2} = \hat{T}\rho_1\hat{T}^{+}. \tag{5}$$

Because of the stimulated scattering, the coefficients of the matrix of transformation $\hat{T}$ can depend on state 2, i.e., are functions of N_2 and $\vec{S}_2$. This relationship can be found explicitly by using the following expansion for $\hat{T}$:

$$\hat{T} = AI + \vec{B}\cdot\vec{\sigma}. \tag{6}$$

After simple algebraic calculations, one gets

$$\left(\frac{\partial \rho_2}{\partial t}\right)_{1\to 2} = \left[\frac{1}{2}(A^2 + B^2)N_1 + 2A(\vec{S}_1\cdot\vec{B})\right]I + [AN_1\vec{B} + (A^2 + B^2)\vec{S}_1]\cdot\vec{\sigma}. \tag{7}$$

It is obvious from the symmetry of the problem that $\vec{B}$ is parallel to $\vec{S}_2$. Comparison of Eqs. (4) and (5) allows one to obtain

$$A^2 + B^2 = W_{12}\left(\frac{N_2}{2} + 1\right), \tag{8}$$

$$2A\vec{B} = W_{12}\vec{S}_2. \tag{9}$$

Using Eqs. (7)–(9) one can express the variation of the density matrix:

$$\left(\frac{\partial \rho_2}{\partial t}\right)_{1\to 2} = \frac{1}{2}W_{12}\left[N_1\left(\frac{N_2}{2}+1\right)+2\vec{S}_1\cdot\vec{S}_2\right] + W_{12}\left[\frac{1}{2}N_1\vec{S}_2 + \left(\frac{N_2}{2}+1\right)\vec{S}_1\right]\vec{\sigma}. \tag{10}$$

Taking into account both incoming and outgoing transitions, one finally obtains the equations for the occupation numbers and the pseudospins:

$$\frac{dN_2}{dt} = (W_{12} - W_{21})\left[\frac{1}{2}N_1N_2 + 2(\vec{S}_1\cdot\vec{S}_2)\right] + (W_{12}N_1 - W_{21}N_2), \tag{11}$$

$$\frac{d\vec{S}_2}{dt} = \frac{1}{2}(W_{12} - W_{21})(N_1\vec{S}_2 + N_2\vec{S}_1) + (W_{12}\vec{S}_1 - W_{21}\vec{S}_2). \tag{12}$$

Once we have obtained a set of equations describing the dynamics of the population and pseudospin for two states, it is easy to generalize it for the case of n states and to take into account both the finite polariton lifetime and the pseudospin rotation induced by the polariton longitudinal-transverse splitting:

$$\frac{dN_{\vec{k}}}{dt} = -\frac{1}{\tau_{\vec{k}}}N_{\vec{k}} + \sum_{\vec{k}'}\left\{(W_{\vec{k}'\vec{k}} - W_{\vec{k}\vec{k}'})\left[\frac{1}{2}N_{\vec{k}}N_{\vec{k}'} + 2(\vec{S}_{\vec{k}}\cdot\vec{S}_{\vec{k}'})\right] + (W_{\vec{k}'\vec{k}}N_{\vec{k}'} - W_{\vec{k}\vec{k}'}N_{\vec{k}})\right\}, \tag{13}$$

$$\frac{d\vec{S}_{\vec{k}}}{dt} = -\frac{1}{\tau_{s\vec{k}}}\vec{S}_{\vec{k}} + \sum_{\vec{k}'}\frac{1}{2}[(W_{\vec{k}'\vec{k}} - W_{\vec{k}\vec{k}'})(N_{\vec{k}}\vec{S}_{\vec{k}'} + N_{\vec{k}'}\vec{S}_{\vec{k}}) + (W_{\vec{k}'\vec{k}}\vec{S}_{\vec{k}'} - W_{\vec{k}\vec{k}'}\vec{S}_{\vec{k}})] + [\vec{\Omega}_{\vec{k}}\times\vec{S}]. \tag{14}$$

The pseudospin lifetime $\tau_{s\vec{k}}^{-1} = \tau_{\vec{k}}^{-1} + \tau_{sl}^{-1}$ was introduced to take into account all the other processes of pseudospin relaxation (characterized by the time τ_{sl}) and the radiative decay of polaritons described by the recombination time. The last term in (14) describes the pseudospin precession around an effective in-plane magnetic field [10,12] determined by exciton longitudinal-transverse splitting. This term is responsible for oscillations of the circular polarization degree of the emitted light. $\vec{\Omega}_{\vec{k}}$ is parallel to the in-plane wave vector for $\vec{k} \parallel \vec{x}$; for other directions of $\vec{k}$ it makes with the x axis twice the same angle as $\vec{k}$ [10]. Important specifics of spin relaxation in microcavities comes from the stimulated scattering of polaritons. While in the case of spontaneous scattering the Maialle mechanism manifests itself in oscillations between $+1$ and -1 exciton-spin states [14], if stimulated scattering dominates, one of two allowed exciton-spin states can be populating much more efficiently than the other one, which may result in the increase of the circular polarization degree of photoluminescence with time.

The general set of kinetic equations (13) and (14) simplifies significantly if the problem has a cylindrical symmetry. In this case the in-plane pseudospin projection $S_{\vec{k}}^{\tau}$ is orthogonal to $\vec{\Omega}_{\vec{k}}$ for each given quantum state. Therefore, we can use instead of Eqs. (13) and (14) a reduced set of equations depending only on magnitudes of polariton wave vectors:

$$\frac{dN_k}{dt} = -\frac{1}{\tau_k}N_k + \sum_{k'}\Big[(\tilde{W}_{k'k} - \tilde{W}_{kk'})\left(\frac{1}{2}N_kN_{k'} + 2S_k^zS_{k'}^z\right) + (\tilde{W}_{k'k}N_{k'} - \tilde{W}_{kk'}N_k) + 2(\bar{W}_{k'k} - \bar{W}_{kk'})S_k^{\tau}S_{k'}^{\tau}\Big], \tag{15}$$

$$\frac{dS_k^z}{dt} = -\frac{1}{\tau_{sk}}S_k^z + \sum_{k'}\Big[\frac{1}{2}(\tilde{W}_{k'k} - \tilde{W}_{kk'})(N_kS_{k'}^z + N_{k'}S_k^z) + (\tilde{W}_{k'k}S_{k'}^z - \tilde{W}_{kk'}S_k^z)\Big] - \Omega_kS_k^{\tau}, \tag{16}$$

$$\frac{dS_k^{\tau}}{dt} = -\frac{1}{\tau_{sk}}S_k^{\tau} + \sum_{k'}\Big[\frac{1}{2}(\bar{W}_{k'k} - \bar{W}_{kk'})N_kS_{k'}^{\tau} + \frac{1}{2}(\tilde{W}_{k'k} - \tilde{W}_{kk'})N_{k'}S_k^{\tau} + (\bar{W}_{k'k}S_{k'}^{\tau} - \tilde{W}_{kk'}S_k^{\tau})\Big] + \Omega_kS_k^z, \tag{17}$$

where $\bar{W}_{kk'} = \frac{1}{2\pi}\int_0^{2\pi} W_{\vec{k}'\vec{k}}\cos 2\varphi\, d\varphi$, $\tilde{W}_{kk'} = \frac{1}{2\pi}\int_0^{2\pi} \times W_{\vec{k}'\vec{k}}d\varphi$, φ is the angle between $\vec{k}$ and $\vec{k}'$, and $N_k = 2\pi k N_{\vec{k}}$.

While numerically solving the Boltzmann equations (15)–(17), we have considered polaritons scattering with acoustic phonons or electrons, the polariton-polariton interaction being neglected. This scattering, although essential for energy relaxation of exciton-polaritons, does not seem to affect strongly the circular polarization degree of emission of microcavities since it conserves the total spin. The parameters of the structure are taken from Ref. [6], and the analytical expressions for matrix elements describing polariton scattering are taken from Ref. [15]. We have assumed the QWs contain a free electron gas of density 10^{11} cm^{-2} at the lattice temperature (10 K).

Figure 3 shows the polarization degree of light emitted by the cavity at zero detuning for three different emission

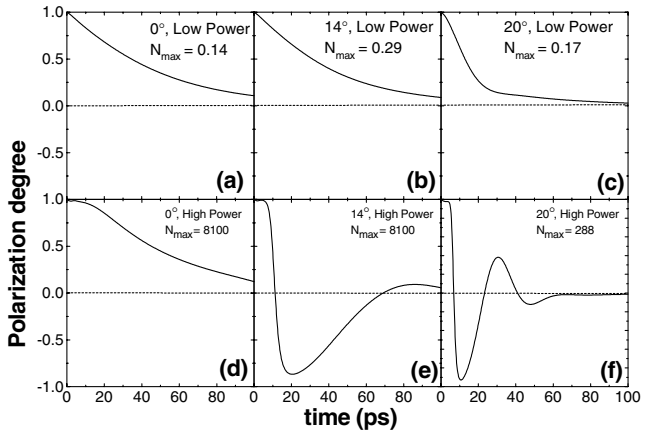

FIG. 3. Polarization degree of the photoluminescence at different angles (0°, 14°, and 20°) from the model CdTe microcavity at zero detuning, calculated using Eqs. (15)–(17) for a pulsed nonresonant excitation with the excitation power of 7μJ per pulse (a)–(c) and excitation power of 700 μJ per pulse (d)–(f). The maximum value polariton occupation numbers achieved at the given angles are indicated on the figures.

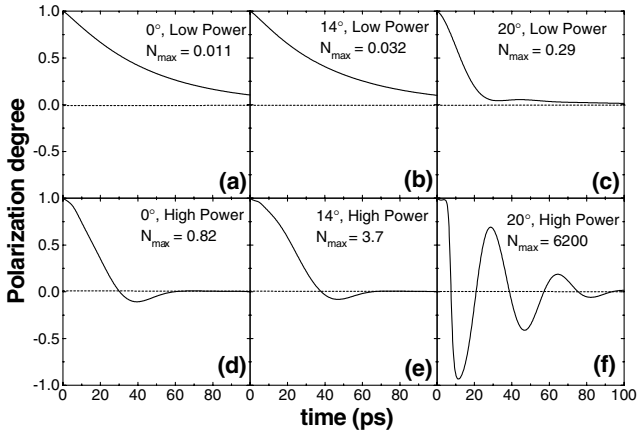

FIG. 4. The same as Fig. 3, but for the negative detuning of -10 meV.

angles. Two different pumping powers, below and above stimulation threshold, are considered. Below threshold, the spin system is in the collision-dominated regime [14] and the polarization degree displays a monotonic decay. The decay is faster for the angle of 20° due to the larger value of the TE-TM splitting at this angle. The situation changes dramatically above the stimulation threshold. The polarization degree oscillates with a period proportional to the inverse of the TE-TM splitting. It is also sensitive to the pumping power due to the interplay between stimulated scattering and spin rotation. For zero detuning between cavity and exciton modes, the polarization degree of the ground state does not oscillate because of the vanishing value of the TE-TM splitting at this point. Figure 4 shows the same as Fig. 3 but for -10 meV detuning, where a strong bottleneck effect arises (i.e., the maximum of polariton population corresponds to a nonzero in-plane wave vector of light). In our case the bottleneck region lies at about 20°.

The polarization degree at the bottleneck is found to influence the polarization degree of all lower states. Indeed, in contrast to the zero detuning case, the polarizations at the ground state and at 14° look similar, both experiencing damped oscillation.

In conclusion, we have developed a formalism describing polariton relaxation in semiconductor microcavities taking into account self-consistently stimulated scattering and spin-relaxation processes induced by the splitting between TE- and TM-polarized exciton-polaritons. The interplay between these two effects leads, in the high excitation regime, to pronounced oscillations in circular polarization of light emitted by the cavity. This demonstrates, in agreement with recent experimental observations, that the conservation of the spin coherence is compatible with fast relaxation processes. This result opens a way to realization of spin-optronics devices based on microcavities.

We thank F. P. Laussy for critical reading of the manuscript and technical help. This work has been supported by the European Research Office of the U.S. Army, Contract No. N62558-03-M0803.

*Permanent address: St. Petersburg State Polytechnical University, 29, Politechnicheskaya, 195251 St. Petersburg, Russia.

[1] A. V. Kavokin and G. Malpuech, *Cavity Polaritons* (Elsevier, Amsterdam, 2003).
[2] H. Deng *et al.*, Science **298**, 199 (2002).
[3] Yu. G. Rubo *et al.*, Phys. Rev. Lett. **91**, 156403 (2003).
[4] P. G. Lagoudakis *et al.*, Phys. Rev. B **65**, 161310 (2002).
[5] A. V. Kavokin *et al.*, Phys. Rev. B **67**, 195321 (2003).
[6] M. D. Martin *et al.*, Phys. Rev. Lett. **89**, 077402 (2002).
[7] R. J. Elliot, Phys. Rev. **96**, 266 (1954); Y. Yafet, *Solid State Physics* (Academic Press, New York, 1963), Vol. 14, p. 1.
[8] G. E. Pikus and G. L. Bir, Zh. Eksp. Teor. Fiz. **60**, 195 (1971) [Sov. Phys. JETP **33**, 108 (1971)].
[9] M. I. D'yakonov and V. I. Perel, Fiz. Tverd. Tela (Leningrad) **13**, 3851 (1971) [Sov. Phys. Solid State **13**, 3023 (1972)].
[10] M. Z. Maialle, D. E. de Andrada e Silva, and L. J. Sham, Phys. Rev. B **47**, 15 776 (1993).
[11] R. I. Dzhioev *et al.*, Phys. Rev. B **56**, 13 405 (1997).
[12] F. Tassone, F. Bassani, and L. C. Andreani, Phys. Rev. B **45**, 6023 (1992).
[13] G. Panzarini *et al.*, Phys. Rev. B **59**, 5082 (1999).
[14] M. A. Brand *et al.*, Phys. Rev. Lett. **89**, 236601 (2002).
[15] G. Malpuech *et al.*, Phys. Rev. B **65**, 153310 (2002).