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Anisotropic polariton scattering and spin dynamics of cavity polaritons

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Abstract

We describe the spin-dynamics of exciton–polaritons in semiconductor microcavities in the strong coupling regime. Using the Liouville equation for the spin-density matrix in the Born–Markov approximation we obtain kinetic equations taking into account polariton–acoustic phonon and polariton–polariton scattering. We describe both the 'polariton laser' regime (non-resonant excitation) and 'optical parametric oscillator' regime (resonant excitation at the magic angle). We obtain a good agreement with experimental data on the dynamics of polarization of light emitted by microcavities.

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1. Introduction

A remarkable peculiarity of exciton–polaritons in microcavities consists in a possibility of final state stimulation of their scattering that makes both their energy and spin relaxation subject to non-trivial bosonic effects. Recent experiments have shown picosecond-scale oscillations in circular polarization degree of emission from microcavities under resonant [1] or non-resonant polarized pumping [2]. Recently we have published a theoretical work [3] presenting the spin-density matrix technique which allowed to describe polariton spin relaxation under non-resonant excitation taking into account polariton coupling with acoustic phonons. We have shown that experimentally observed oscillations of the polarization degree [2] are linked to the beats between linearly polarized TE and TM polariton modes. Final state stimulation of polariton scattering has an important effect on the spin dynamics as well allowing for conservation and even amplification of the polarization degree.

In the present work we present a new, more general formalism accounting for polariton–polariton scattering as well and allowing to describe also the 'optical parametric oscillator' regime [1] where resonant polariton–polariton scattering dominates over other relaxation mechanisms.

2. Formalism

Our starting point is the Liouville equation for the complete density matrix of the system which in the interaction representation reads

$$i\hbar\frac{d\rho}{dt} = [\hat{H}(t), \rho] \tag{1}$$

where the time-dependent Hamiltonian of the system describing polariton–phonon and polariton–polariton interaction is as follows

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$$\hat{H} = \sum_{\mathbf{k},\mathbf{k}'} V_{\mathbf{k}k'}\, e^{i(\Omega_{k'}-\Omega_{\mathbf{k}}-\omega_{k'-k})t} b_{\mathbf{k}'-\mathbf{k}}(a_{\mathbf{k}\uparrow}a^{+}_{\mathbf{k}'\uparrow} + a_{\mathbf{k}\downarrow}a^{+}_{\mathbf{k}'\downarrow})$$
$$+ \sum_{\mathbf{k},\mathbf{k}',\mathbf{k}''} [V^{(1)}_{\mathbf{k};\mathbf{k}';\mathbf{k}'';\mathbf{k}+\mathbf{k}'-\mathbf{k}''}\, e^{i(\Omega_{k'}+\Omega_{k+k'-k''}-\Omega_{k}-\Omega_{k'})t}$$
$$\times(a_{\mathbf{k}\uparrow}a_{\mathbf{k}'\uparrow}a^{+}_{\mathbf{k}''\uparrow}a^{+}_{\mathbf{k}+\mathbf{k}'-\mathbf{k}''\uparrow} + a_{\mathbf{k}\downarrow}a_{\mathbf{k}'\downarrow}a^{+}_{\mathbf{k}''\downarrow}a^{+}_{\mathbf{k}+\mathbf{k}'-\mathbf{k}''\downarrow}) \quad (2)$$
$$+V^{(2)}_{\mathbf{k};\mathbf{k}';\mathbf{k}'';\mathbf{k}+\mathbf{k}'-\mathbf{k}''}\, e^{i(\Omega_{k'}+\Omega_{k+k'-k''}-\Omega_{k}-\Omega_{k'})t}$$
$$\times(a_{\mathbf{k}\uparrow}a_{\mathbf{k}'\downarrow} + a_{\mathbf{k}\downarrow}a_{\mathbf{k}'\uparrow})$$
$$\times(a^{+}_{\mathbf{k}''\uparrow}a^{+}_{\mathbf{k}+\mathbf{k}'-\mathbf{k}''\downarrow} + a^{+}_{\mathbf{k}''\downarrow}a^{+}_{\mathbf{k}+\mathbf{k}'-\mathbf{k}''\uparrow})] + \text{h.c.}$$

In (2) $a_{\mathbf{k}\uparrow}(a^{+}_{\mathbf{k}\uparrow})$ and $a_{\mathbf{k}\downarrow}(a^{+}_{\mathbf{k}\downarrow})$ are the annihilation (creation) operators for the polaritons with wave-vector $\mathbf{k}$ having the spin projection ±1 on the structure growth axis, $b_{\mathbf{q}}$ and $b^{+}_{\mathbf{q}}$ are operators for phonons with wave-vector $\mathbf{q}$, $\Omega_{\mathbf{k}}$ and $\omega_{\mathbf{q}}$ are the polariton and phonon dispersions, respectively. We do not consider here the dark exciton states with spin projections equal to ±2. The first term in (2) proportional to $V_{\mathbf{kk}'}$ corresponds to the scattering of polaritons with acoustic phonons, the second and third ones describe the polariton–polariton scattering which is characterized by two matrix elements: $V^{(1)}_{\mathbf{k};\mathbf{k}';\mathbf{k}'';\mathbf{k}'''}$ and $V^{(2)}_{\mathbf{k};\mathbf{k}';\mathbf{k}'';\mathbf{k}'''}$ for the scattering of the polaritons with the same and opposite spin projections, respectively. In the case of dominant exchange [4], $V^{(1)}_{\mathbf{k};\mathbf{k}';\mathbf{k}'';\mathbf{k}'''} \gg V^{(2)}_{\mathbf{k};\mathbf{k}';\mathbf{k}'';\mathbf{k}'''}$, however, if scattering goes also via bipolariton resonance one can have these matrix elements being of the same order of magnitude. This may be the case near the 'magic' angle where polariton and bipolariton dispersion curves cross each other [5]. Note, that if $V^{(1)}_{\mathbf{k};\mathbf{k}';\mathbf{k}'';\mathbf{k}'''} \neq 2V^{(2)}_{\mathbf{k};\mathbf{k}';\mathbf{k}'';\mathbf{k}'''}$ the polariton–polariton scattering is anisotropic, i.e. its intensity depends on the absolute orientation of the spins.

Integrating the Liouville equation and substituting the obtained expression for the density matrix into the right hand side of it, one can rewrite (1) using Markov approximation [8] in the following form:

$$\dot{\rho}(t) = -\frac{1}{\hbar^2}\int_{-\infty}^{t} [\hat{H}(t), [\hat{H}(\tau), \rho(t)]]\mathrm{d}\tau \quad (3)$$

The complete density matrix is treated in the Born approximation and is factorized into the product of the phonon density matrix and polariton density matrices corresponding to the different states in the reciprocal space

$$\rho = \rho_{\mathrm{ph}} \otimes \prod_{\mathbf{k}} \rho_{\mathbf{k}} \quad (4)$$

where $\rho_{\mathbf{k}}$ is the spin-density matrix for the polaritons with the wave-vector $\mathbf{k}$. The diagonal elements of this density matrix give the populations of the spin-up and spin-down states: $N_{\mathbf{k},\uparrow} = \mathrm{Tr}(a^{+}_{\mathbf{k}\uparrow}a_{\mathbf{k}\uparrow}\rho_{\mathbf{k}}), N_{\mathbf{k},\downarrow} = \mathrm{Tr}(a^{+}_{\mathbf{k},\downarrow}a_{\mathbf{k},\downarrow}\rho_{\mathbf{k}})$, while off-diagonal components are connected with the in-plane projection of the polariton pseudospin [3] $S_{\perp,\mathbf{k}} = S_{x,\mathbf{k}}\mathbf{e}_x + S_{y,\mathbf{k}}\mathbf{e}_y$, in the following way $S_{x,\mathbf{k}} - \mathrm{i}S_{y,\mathbf{k}} = \mathrm{Tr}(a^{+}_{\mathbf{k}\uparrow}a_{\mathbf{k}\downarrow}\rho_{\mathbf{k}})$. Here $\mathbf{e}_x$ and $\mathbf{e}_y$ are the unit vectors in the cavity plane.

These two approximations allow to perform of the time-integration in (3), which yields the energy conservation rule for the scattering acts and allows to 'trace out' the phonon part of the density matrix. Finally, a Lindblad-type equation [9] for the polariton density matrix can be obtained, which allows the derivation of the kinetic equations for the polariton occupation numbers and in-plane projections of their pseudospins. Simple, but rather tiresome algebraic calculations give

$$\frac{\mathrm{d}N_{\mathbf{k}\uparrow}}{\mathrm{d}t} = \sum_{\mathbf{k}'}\{W_{\mathbf{k}'\to\mathbf{k}}[N_{\mathbf{k}',\uparrow}(N_{\mathbf{k},\uparrow}+1) + (\mathbf{S}_{\perp,\mathbf{k}}\cdot\mathbf{S}_{\perp,\mathbf{k}'})]$$
$$- W_{\mathbf{k}\to\mathbf{k}'}[N_{\mathbf{k},\uparrow}(N_{\mathbf{k}',\uparrow}+1) + (\mathbf{S}_{\perp,\mathbf{k}}\cdot\mathbf{S}_{\perp,\mathbf{k}'})]\}$$
$$+ \sum_{\mathbf{k}',\mathbf{k}''}\{W^{(1)}_{\mathbf{k},\mathbf{k}',\mathbf{k}'',\mathbf{k}+\mathbf{k}'-\mathbf{k}''}[(N_{\mathbf{k}\uparrow}+N_{\mathbf{k}'\uparrow}$$
$$+1)N_{\mathbf{k}''\uparrow}N_{\mathbf{k}+\mathbf{k}'-\mathbf{k}''\uparrow} - (2N_{\mathbf{k}''\uparrow}+1)N_{\mathbf{k}\uparrow}N_{\mathbf{k}'\uparrow}]$$
$$+ 2W^{(2)}_{\mathbf{k},\mathbf{k}',\mathbf{k}'',\mathbf{k}+\mathbf{k}'-\mathbf{k}''}[(N_{\mathbf{k}\uparrow}+N_{\mathbf{k}'\downarrow}+1)$$
$$\times(N_{\mathbf{k}''\uparrow}N_{\mathbf{k}+\mathbf{k}'-\mathbf{k}''\downarrow} + \mathbf{S}_{\perp,\mathbf{k}''}\mathbf{S}_{\perp,\mathbf{k}+\mathbf{k}'-\mathbf{k}''}) \quad (5)$$
$$- (N_{\mathbf{k}\uparrow}N_{\mathbf{k}'\downarrow} + \mathbf{S}_{\perp,\mathbf{k}}\mathbf{S}_{\perp,\mathbf{k}'})(N_{\mathbf{k}''\uparrow}+N_{\mathbf{k}''\downarrow}+1)]$$
$$+ 4W^{(12)}_{\mathbf{k},\mathbf{k}',\mathbf{k}'',\mathbf{k}+\mathbf{k}'-\mathbf{k}''}[(N_{\mathbf{k}''\uparrow}(\mathbf{S}_{\perp,\mathbf{k}'}\mathbf{S}_{\perp,\mathbf{k}+\mathbf{k}'-\mathbf{k}''})$$
$$- N_{\mathbf{k}\uparrow}(\mathbf{S}_{\perp,\mathbf{k}'}\mathbf{S}_{\perp,\mathbf{k}+\mathbf{k}'-\mathbf{k}''})]$$
$$+ 2W^{(12)}_{\mathbf{k},\mathbf{k}',\mathbf{k}'',\mathbf{k}+\mathbf{k}'-\mathbf{k}''}(\mathbf{S}_{\perp,\mathbf{k}}\mathbf{S}_{\perp,\mathbf{k}+\mathbf{k}'-\mathbf{k}''})$$
$$\times(N_{\mathbf{k}''\uparrow}+N_{\mathbf{k}''\downarrow}-N_{\mathbf{k}'\uparrow}+N_{\mathbf{k}'\downarrow})$$

$$\frac{\mathrm{d}\mathbf{S}_{\perp,\mathbf{k}}}{\mathrm{d}t} = \frac{1}{2}\sum_{\mathbf{k}'}\{(W_{\mathbf{k}'\to\mathbf{k}} - W_{\mathbf{k}\to\mathbf{k}'})[(N_{\mathbf{k}'\uparrow}+N_{\mathbf{k}'\downarrow})\mathbf{S}_{\perp,\mathbf{k}}$$
$$+(N_{\mathbf{k}\uparrow}+N_{\mathbf{k}\downarrow})\mathbf{S}_{\perp,\mathbf{k}'}] + 2W_{\mathbf{k}'\to\mathbf{k}}\mathbf{S}_{\perp,\mathbf{k}'} - 2W_{\mathbf{k}\to\mathbf{k}'}\mathbf{S}_{\perp,\mathbf{k}}\}$$
$$+ \sum_{\mathbf{k}',\mathbf{k}''}\left\{\frac{W^{(1)}_{\mathbf{k},\mathbf{k}',\mathbf{k}'',\mathbf{k}+\mathbf{k}'-\mathbf{k}'}}{2}\mathbf{S}_{\perp,\mathbf{k}}[N_{\mathbf{k}''\uparrow}N_{\mathbf{k}+\mathbf{k}'-\mathbf{k}''\uparrow}\right.$$
$$+ N_{\mathbf{k}''\downarrow}N_{\mathbf{k}+\mathbf{k}'-\mathbf{k}''\downarrow} - N_{\mathbf{k}'\uparrow}(2N_{\mathbf{k}''\uparrow}+1) - N_{\mathbf{k}'\downarrow}(2N_{\mathbf{k}''\downarrow}$$
$$+1)] + W^{(1)}_{\mathbf{k},\mathbf{k}',\mathbf{k}'',\mathbf{k}+\mathbf{k}'-\mathbf{k}''}(2\mathbf{S}_{\perp,\mathbf{k}''}(\mathbf{S}_{\perp,\mathbf{k}'}\cdot\mathbf{S}_{\perp,\mathbf{k}+\mathbf{k}'-\mathbf{k}''})$$
$$- \mathbf{S}_{\perp,\mathbf{k}'}(\mathbf{S}_{\perp,\mathbf{k}''}\cdot\mathbf{S}_{\perp,\mathbf{k}+\mathbf{k}'-\mathbf{k}''})) \quad (6)$$
$$+ W^{(2)}_{\mathbf{k},\mathbf{k}',\mathbf{k}'',\mathbf{k}+\mathbf{k}'-\mathbf{k}''}[2(\mathbf{S}_{\perp,\mathbf{k}}+\mathbf{S}_{\perp,\mathbf{k}'})(N_{\mathbf{k}''\uparrow}N_{\mathbf{k}+\mathbf{k}'-\mathbf{k}''\downarrow}$$
$$+ (\mathbf{S}_{\perp,\mathbf{k}''}\cdot\mathbf{S}_{\perp,\mathbf{k}+\mathbf{k}'-\mathbf{k}''})) - (\mathbf{S}_{\perp,\mathbf{k}}(N_{\mathbf{k}'\uparrow}+N_{\mathbf{k}'\downarrow})$$
$$+ (\mathbf{S}_{\perp,\mathbf{k}'}(N_{\mathbf{k}\uparrow}+N_{\mathbf{k}\downarrow}))(N_{\mathbf{k}''\uparrow}+N_{\mathbf{k}''\downarrow}+1)]$$
$$- 4W^{(12)}_{\mathbf{k},\mathbf{k}',\mathbf{k}'',\mathbf{k}+\mathbf{k}'-\mathbf{k}''}\mathbf{S}_{\perp,\mathbf{k}}(\mathbf{S}_{\perp,\mathbf{k}'}\cdot\mathbf{S}_{\perp,\mathbf{k}''})$$
$$+ W^{(12)}_{\mathbf{k},\mathbf{k}',\mathbf{k}'',\mathbf{k}+\mathbf{k}'-\mathbf{k}''}\mathbf{S}_{\perp,\mathbf{k}+\mathbf{k}'-\mathbf{k}''}[2((N_{\mathbf{k}'\uparrow}+1)N_{\mathbf{k}''\uparrow}$$
$$+ (N_{\mathbf{k}'\downarrow}+1)N_{\mathbf{k}''\downarrow}) + (N_{\mathbf{k}''\uparrow}+N_{\mathbf{k}''\downarrow}-N_{\mathbf{k}'\uparrow}-N_{\mathbf{k}'\downarrow})$$
$$\times(N_{\mathbf{k}\uparrow}+N_{\mathbf{k}\downarrow})]$$

where the transition rates for polariton–phonon scattering are as follows

$$W_{\mathbf{k}\to\mathbf{k}'} = \frac{2}{\hbar\gamma}(n_{\mathrm{ph},\mathbf{k}-\mathbf{k}'}+1)|V_{\mathbf{k}\mathbf{k}'}|^2, \quad W_{\mathbf{k}'\to\mathbf{k}} = \frac{2\pi}{\hbar\gamma} n_{\mathrm{ph},\mathbf{k}-\mathbf{k}'}|V_{\mathbf{k}\mathbf{k}'}|^2$$

where γ is a radiative broadening of the states $\mathbf{k}$ and $\mathbf{k}'$, $n_{\mathrm{ph},\mathbf{q}}$ is the number of phonons in the state $\mathbf{q}$, and polariton–polariton transition rates are

$$W^{(i)}_{\mathbf{k},\mathbf{k}',\mathbf{k}'',\mathbf{k}'''} = \frac{2}{\hbar\gamma}|V^{(i)}_{\mathbf{k},\mathbf{k}',\mathbf{k}'',\mathbf{k}'''}|^2,$$

$$W^{(12)}_{\mathbf{k},\mathbf{k}',\mathbf{k}'',\mathbf{k}'''} = \frac{2}{\hbar\gamma}\mathrm{Re}(V^{(1)}_{\mathbf{k},\mathbf{k}',\mathbf{k}'',\mathbf{k}'''}V^{*(2)}_{\mathbf{k},\mathbf{k}',\mathbf{k}'',\mathbf{k}'''})$$

The equation for spin-down occupation numbers can be obtained from (5) by changing the spin indices.

If only polariton–acoustic phonon interaction is present, Eqs. (5) and (6) coincide with those obtained using the method of the linear transformation of spin density matrix [3]. The system (5) and (6) describes the population and spin dynamics of exciton–polaritons at any ratio of singlet and triplet scattering elements and generalizes previous works [3,6].

3. Results and discussion

The system of the kinetic Eqs. (5) and (6) is rather complicated and its numerical treatment for the general case of non-resonant excitation represents an extremely difficult computational task. Here we apply the general formalism developed above to the resonant excitation case, when a microcavity is pumped at the so-called magic angle [1]. In this configuration the resonant scattering the polariton pairs excited by the pump pulse toward the signal and the idler state is the dominant relaxation process. The intensity and polarization of the emission could be governed by the additional weak probe pulse, sent to the $\boldsymbol{k}=0$ (signal) state. The polarization dynamics of such an optical parametric oscillator (OPO) was studied experimentally in the case of circularly polarized probe by Lagoudakis et al. [1] and theoretically in [6]. Here we consider the case of linearly polarized probe.

In the regime of parametric amplification, only three states (pump, signal and idler) govern the polariton relaxation dynamics. Therefore, the system of kinetic Eqs. (5) and (6) can be reduced to 9 equations describing the dynamics of occupation numbers and in-plane components of the pseudospins of these three states. The intensities of the circularly polarized components of the signal emission are proportional to the number of polaritons in the spin-up and spin-down states, $I_{\sigma^{+,-}} \propto N_{\uparrow,\downarrow}$, the intensities of the horizontally and vertically polarized components are $I_{\leftrightarrow,\updownarrow} \propto 1/2(N_\uparrow + N_\downarrow) \pm S_x$ and the intensities of the diagonal components are $I_{\swarrow,\nwarrow} \propto 1/2(N_\uparrow + N_\downarrow) \pm S_y$. The polariton–acoustic phonon interactions can be completely excluded from our consideration, since they are dominated by polariton–polariton scattering in the OPO regime. We have taken into account the effective magnetic field which arises from the polariton–polariton interactions if $N_\uparrow \neq N_\downarrow$ [6,7].

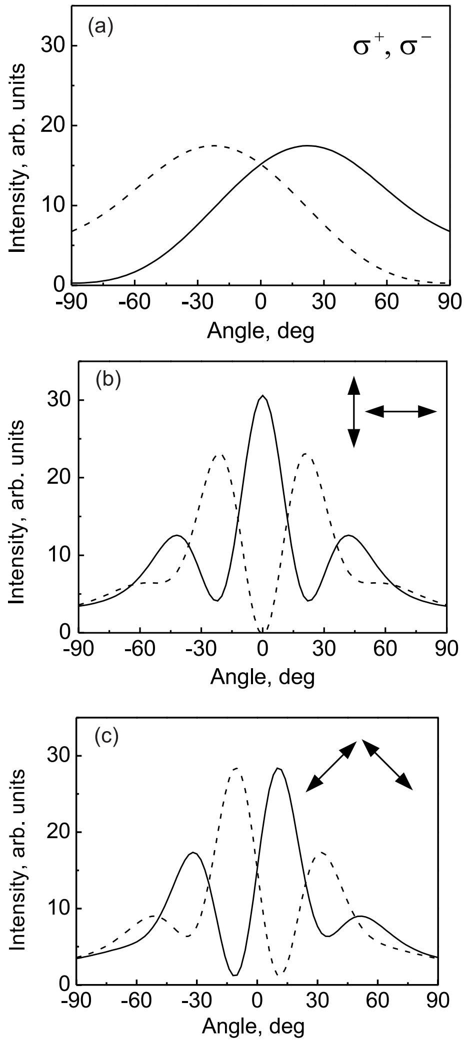

Fig. 1. Calculated intensity of signal in different polarizations versus angle between pump pseudopsin and x-axis. Pump polarization changes from σ^- to σ^+ passing through elliptical and horizontal linear polarization (parallel to x-axis at zero angle). The initial pump population is 10^5. The probe is linearly polarized along x-axis, its initial population is 200. (a) Intensities of σ^+ (solid) and σ^- (dashed) circular components. (b) Intensities of horizontal (solid) and vertical (dashed) linear polarizations. (c) Intensities of diagonal linear polarization components.

The results of numerical simulation are shown at Fig. 1. One can see that if the pump and probe are co-linearly polarized the signal emission in the same polarization is strongly enhanced as compared to the cases of elliptical and circular pump. In the case of elliptically polarized pump pulse, the signal also becomes elliptical while the direction of the main axis of the ellipse rotates as a function of the circular polarization degree of the pump like in [1].

In conclusion, we have developed a general formalism allowing to obtain the kinetic equations for the cavity exciton–polaritons fully taking into account their polarization and anisotropy of polariton–polariton interactions. This method allows to describe polariton dynamics in under resonant or non-resonant excitation.

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