Optical orientation of the homogeneous nonequilibrium Bose-Einstein condensate of exciton polaritons

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A simple model, describing the steady state of the nonequilibrium polarization of a homogeneous Bose-Einstein condensate of exciton polaritons, is considered. It explains the suppression of spin splitting of a nonequilibrium polariton condensate in an external magnetic field, the linear polarization, the linear-to-circular polarization conversion, and the unexpected sign of the circular polarization of the condensate all on equal footing. It is shown that inverse effects are possible, to wit, spontaneous circular polarization and the enhancement of spin splitting of a nonequilibrium condensate of polaritons.

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I. INTRODUCTION

Artificially created systems of bosons are attractive for studying the different aspects of Bose-Einstein condensation.[1] The condensates of indirect excitons in quantum wells[2] and bright excitons (polaritons) in microcavities[3,4] are unique, since their properties can be controlled optically. In spite of the nonequilibrium character of optical creation, a quasiequilibrium polariton condensate still can be formed close to the momentum $k = 0$.[4] The states of excitons and polaritons are characterized by projections of the angular momentum ± 1 on the growth axis of the well and can be described in terms of a fictive spin or pseudospin 1/2.[5] Recently the suppression of spin splitting of a polariton condensate has been observed in a steady-state regime.[6] The sign of circular polarization of the condensate appeared to be opposite to that of the equilibrium condensate considered previously in Ref. 7. In turn, the short lifetime of polaritons (a few picoseconds) points to the strong deviation of the spin system from equilibrium. Only the spin dynamics of a nonequilibrium condensate was considered up to now.[8] However a self-consistent explanation of the experiment[6] is absent.

Here a simple model of nonequilibrium spin polarization of a homogeneous ensemble of polaritons is considered in the steady-state regime. This model makes it possible to describe all the principal experimental results, such as the suppression of Zeeman splitting of a nonequilibrium condensate and the unexpected sign of the circular polarization of polaritons within a solely nonequilibrium approach. It predicts also the inverse effects: the spontaneous circular polarization and the enhancement of spin splitting of a nonequilibrium condensate of polaritons in a zero external magnetic field.

II. BRIGHT EXCITON AS A QUASIPARTICLE WITH PSEUDOSPIN 1/2

A model of the pseudospin 1/2 for an ensemble of isolated bright excitons was worked out in Refs. 5 and 9. The two states of a bright (optically allowed) exciton in a GaAs-type quantum well can be considered as the states of a quasiparticle with pseudospin 1/2. It is natural to choose the states with momentum projection $|+1\rangle$ and $|-1\rangle$ on the growth axis of the well, as a projection of the pseudospin "up" $|z,+1/2\rangle$ and "down" $|z,-1/2\rangle$, respectively, on the z axis in the effective three-dimensional space of the pseudospin (x,y,z) (one should not confuse the axes of pseudospin space with the axes in the real space). In the state $|+1\rangle$ ($|-1\rangle$) the exciton emits circularly polarized light in the $\sigma^+(\sigma^-)$ polarization. Then the z component S_z of the mean (over the ensemble of excitons) pseudospin determines the degree of circular polarization of the luminescence, $P_c = 2S_z$. The state $|X\rangle(|Y\rangle)$, which is dipole active along the crystallographic axis $[1\bar{1}0]$([110]), is described by the x component of the pseudospin $|x,+1/2\rangle(|x,-1/2\rangle)$. The x projection of the mean pseudospin determines the degree of linear polarization of the exciton luminescence relative to the crystal axes $[1\bar{1}0]$ and [110]: $P_l = 2S_x$. Finally, the state $|X'\rangle(|Y'\rangle)$, which is polarized along the direction [100] ([010]), corresponds to the projection of the pseudospin $|y,+1/2\rangle(|y,-1/2\rangle)$ on the y axis. The y component of the mean pseudospin determines the degree of linear polarization of the exciton luminescence relative to the crystallographic axes [100] and [010]: $P_{l'} = 2S_y$. All three components of the mean pseudospin can be measured, since they are connected unambiguously with three Stokes parameters characterizing the luminescence polarization:

$$P_c = 2S_z; \quad P_l = 2S_x; \quad P_{l'} = 2S_y. \tag{1}$$

Relations (1) between the light polarization and pseudospin admit a simple geometric interpretation. As is known, the light polarization is characterized by three Stokes parameters, which can also be considered as a vector with coordinates $\vec{P} = (P_l, P_{l'}, P_c)$ on the Poincaré sphere. The pseudospin of an ensemble of excitons can be conceived as a vector $\vec{S}$ in the three-dimensional space. By virtue of (1), each position of pseudospin in the effective three-dimensional space corresponds here to a definite position of vector $\vec{P}$ on the Poincaré sphere.

The Hamiltonian of excitons describes the time evolution of exciton polarization. In the case of symmetry C_{2v} (e.g., an asymmetric well, grown in the direction [001]) the main axes of anisotropic electron-hole exchange interaction in an exciton coincide with the crystal axes [110] and $[1\bar{1}0]$. The Hamiltonian of a bright exciton in a magnetic field in Faraday geometry $\vec{B} \parallel [001]$ can be written in terms of pseudospin as[5]

$$\hat{H} = \frac{\hbar}{2}(\omega_b \hat{\sigma}_x + \Omega_{\text{ext}} \hat{\sigma}_z) = \frac{\hbar}{2}\vec{\Omega} \cdot \vec{\sigma}, \tag{2}$$

where σ_i are Pauli matrices on the x,y,z axes of the pseudospin effective space, and $\Omega_{\rm ext} = \mu_B g_{||} B/\hbar$ is the Larmor precession frequency of the pseudospin in an external field ($g_{||}$ is the longitudinal g factor of the exciton). In the absence of magnetic field ($\Omega_{\rm ext} = 0$) the components of the radiative doublet are polarized along the axes $[1\bar{1}0]$ and $[110]$ with the energy of splitting $\delta_b = \hbar\omega_b$. The pseudospin dynamics is described by the Bloch equation[9]

$$\frac{d\vec{S}}{dt} = \frac{\vec{S}^0 - \vec{S}}{\tau_b} + \vec{\Omega} \times \vec{S}, \tag{3}$$

where on the right side the first term describes the generation and recombination of the pseudospin, and the second term Larmor precession with frequency $\vec{\Omega} = (\omega_b, 0, \Omega_{\rm ext})$. If the lifetime τ_b of the exciton is sufficiently long, so that $\Omega\tau_b \gg 1$, then the Larmor precession about the $\vec{\Omega}$ vector depolarizes the components of the initial pseudospin $\vec{S}^0 = (S_x^0, S_y^0, S_z^0)$ transverse to $\vec{\Omega}$, while the component along $\vec{\Omega}$ is conserved. As a result, the stationary orientation of the pseudospin is attained by projection of $\vec{S}^0$ onto the direction of $\vec{\Omega}$. That simple model explains naturally all polarization phenomena in the ensemble of isolated excitons localized in various nanostructures.[5,10–12]

III. EXCITON-EXCITON INTERACTIONS WITHIN THE PSEUDOSPIN-1/2 MODEL

Even in a diluted system of excitons with concentration $N_x \ll 1/\pi a_x^2$ (within the circle of the Bohr radius of the exciton a_x there are no other excitons[13]) there occurs their spin-spin interaction with each other. It has been found[14] that during creation of excitons in a quantum well by circularly polarized light, the luminescence spectrum is split into two lines, corresponding to recombination of excitons from states $|+1\rangle$ and $|-1\rangle$. This splitting has been explained as due to the exciton-exciton interaction. This interaction can be interpreted as an effective exchange interaction of quasiparticles with pseudospin 1/2.[15] In the mean-field approximation (MFA) the spin splitting of an exciton results from the action of an effective exchange magnetic field along z, and proportional to mean pseudospin $S_z = \frac{1}{2}\langle\sigma_z\rangle$ of an exciton ensemble. The MFA spin Hamiltonian of a given exciton takes the form

$$\hat{H}_{\rm MFA} = -\frac{J}{2}\hat{\sigma}_z S_z$$

where J is the exchange constant. The evolution of exciton polarization under the action of the Hamiltonian $H_{\rm MFA}$ can be considered as a precession $\vec{S}$ with angular frequency $\vec{\Omega}'(\vec{S}) = (0, 0, -\omega_{\rm exch} S_z)$, where $\omega_{\rm exch} = J/\hbar$. Generalization of the precession dynamics for the case of interaction between excitons consists in the addition of the term $\vec{\Omega}'(\vec{S}) \times \vec{S}$. Then the total Larmor frequency of the pseudospin,[15]

$$\vec{\Omega} = (\omega_b, 0, \Omega_{\rm ext} - \omega_{\rm exch} S_z), \tag{4}$$

depends on the orientation of the exciton ensemble, thus making the system nonlinear.

IV. OPTICAL ORIENTATION OF SPINOR BOSE-EINSTEIN CONDENSATE

The diluted gas of bright excitons forms a set of quasiparticles with pseudospin 1/2, which obey Bose statistics (the projection of their angular momentum ± 1 is integer). The Bose statistics of excitons will manifest itself at fairly low temperatures, when the de Broglie wavelength $\lambda_{\rm Br}$ of excitons becomes comparable with the characteristic distance between them, $N_x \pi \lambda_{\rm Br}^2 \geqslant 1$ (under fulfillment of the condition of dilution, $N_x \pi a_x^2 \ll 1$). This situation takes place in the system of indirect excitons[2] and in semiconductor microcavities[3,4] with quantum wells. In the last case, under conditions of strong coupling of photons and excitons there arises a hybrid mode, a polariton with momentum projections $+1$ and -1 on the growth axis. Of course, one can also treat the polariton as a quasiparticle with pseudospin 1/2 and apply the above considerations to the ensemble of polaritons. The pseudospin concept was successfully applied to cavity polaritons in Ref. 16 using the exchange term $H_{\rm MFA}$ to describe the effect of self-induced Larmor precession of the polariton pseudospin.[17] At a temperature below the critical T_c the Bose condensation of polaritons occurs,[3] and in such a case the order parameter will be not a scalar, but a vector function, defining the polarization of the condensate.[4] The complex order parameter also can be expressed through the density components of the pseudospin $\vec{S}$ of the condensate. In spite of the nonequilibrium character of optical creation of an ensemble, a quasiequilibrium polariton condensate can be formed close to the momentum $k = 0$.[4] However, due to the very short lifetime of polaritons (a few picoseconds) the spin degree of freedom is essentially nonequilibrium. It was shown[8] that the pseudospin dynamics $\vec{S}$ of a nonequilibrium polariton condensate is still determined by the precession equation (3) in which the vector $\vec{\Omega}(\vec{S})$ is given by Eq. (4), with the quantities in Eq. (4) having similar meaning as for the case of excitons. The dynamic equations (3) and (4) were solved in Ref. 8.

Here I solve Eqs. (3) and (4) in the steady-state regime in the limit $\Omega\tau_b \gg 1$ and show that the nonequilibrium model reproduces all the main experimental phenomena.[6] Only the pseudospin projection onto the direction $\vec{\Omega}$ remains in the stationary state, $\vec{S} = (\vec{\Omega}\vec{S}^0)\vec{\Omega}/\Omega^2$.[18] Let the initial pseudospin of an ensemble of polaritons $\vec{S}^0$ be directed along the x axis [Fig. 1(a)]. It is seen from Fig. 1 that the vector $\vec{\Omega}$ lies in the plane (x,z) and forms an angle $\arctan([\Omega_{\rm ext} - \omega_{\rm exch} S_z]/\omega_b)$ with the x axis. The circular polarization of polaritons $s_z = S_z/S^0 = P_c/2S^0$ [in the functional form $\Omega_{\rm ext}(S_z)$] reads

$$\frac{\Omega_{\rm ext}}{\omega_b} = a s_z + \frac{1}{2s_z}\left(1 \pm \sqrt{1 - (2s_z)^2}\right), \tag{5}$$

where the only parameter of the model, $a = \omega_{\rm exch} S^0/\omega_b$, characterizes the strength of exchange coupling. The dependence $P_c(\Omega_{\rm ext})$ in units of $2|S^0|$ is plotted in Fig. 1(d) for $a = +3$. Using Eq. (5) one can calculate [Fig. 1(c)] the linear polarization P_ℓ determined by the x component of the pseudospin,

$$s_x(\Omega_{\rm ext}) = \frac{S_x}{S^0} = \frac{P_\ell}{2S^0} = \frac{1}{1 + [(\Omega_{\rm ext}/\omega_b) - a s_z]^2}. \tag{6}$$

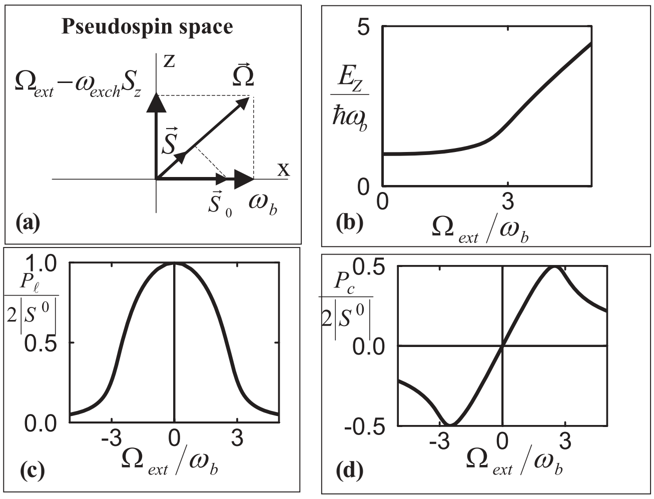

FIG. 1. (a) Orientation of the pseudospin vector under stationary conditions in an external magnetic field ($\Omega_{\text{ext}} \neq 0$) and in the effective magnetic fields of anisotropic electron-hole coupling in an exciton ($\omega_b \neq 0$) and of interparticle spin-spin exchange ($\omega_{\text{exch}} \neq 0$). (b) The Zeeman splitting of the levels $\pm 1/2$ (i.e., the states with angular momentum projection ± 1) vs Ω_{ext} frequency. The dependences of the degrees of linear (c) and circular (d) polarization of polariton luminescence on the Ω_{ext} frequency.

Finally the Zeeman splitting of the pseudospin sublevels,

$$E_Z(\Omega_{\text{ext}}) = \hbar\sqrt{\omega_b^2 + (\Omega_{\text{ext}} - \omega_{\text{exch}} S_z)^2}, \tag{7}$$

is shown in Fig. 1(b). It can be seen that a longitudinal magnetic field $\Omega_{\text{ext}} \neq 0$ brings about the following effects: (i) the exchange interaction of polaritons compensates the Zeeman splitting in an external magnetic field up to $\Omega_{\text{ext}} < 3\omega_b$ [Fig. 1(b)]; (ii) the linear polarization of polaritons S_x decreases [Fig. 1(c)]; (iii) there appears a circular polarization of polaritons, i.e., $S_z \neq 0$ [Fig. 1(d)]; (iv) the sign of the circular polarization of polaritons, S_z, coincides with the sign of Ω_{ext}. All these features were observed experimentally.[6]

V. DISCUSSION

Earlier the items (i)–(iii) were predicted for the spin equilibrium condensate. The authors of Ref. 7 calculated the zero-temperature equilibrium spin $\vec{S}_T = -\frac{1}{2}\frac{\vec{\Omega}}{\Omega}$ where $\vec{\Omega}(\vec{S}_T)$ is given by Eq. (4). According to Ref. 7, however, the sign of the equilibrium circular polarization, S_{Tz} [item (iv)], must be opposite to the sign of Ω_{ext} for *any* Ω_{ext} in clear contradiction with experiment.[6,19] Moreover, the short lifetime (a few picoseconds) of cavity polaritons points to the fact that the spin system should be considered as strongly nonequilibrium from the very outset. Therefore the equilibrium approach[7] cannot provide a self-consistent description of the experiment.

It should be noted that the suppression of spin splitting is not complete ($E_Z \geqslant \hbar\omega_b$), in apparent contradiction with experiment.[6] The fact is that the measurements of splitting were carried out in crossed circular polarizers. In a weak magnetic field, however, two orthogonal spin states are polarized elliptically. Strictly speaking, the photoluminescence (PL) polarization analyzers should also be elliptical, but this would be inconvenient from the practical point of view. Nevertheless, the question of nonzero splitting is central, since an explanation has recently appeared[20] predicting the complete quenching of splitting, i.e., $E_Z = 0$. The given model can be checked easily in a zero field, in which two states of the condensate are linearly polarized orthogonally. Therefore the PL spectra of the condensate must show splitting of $E_Z = \hbar\omega_b$ in two crossed linear polarizations.

Note that for explanation of the experimental results[6] on polarization and quenching of Zeeman splitting, the exchange constant should be ferromagnetic, $J > 0$. Indeed, the Larmor precession frequency in the exchange field is determined by the product JS_z which should be positive for compensation [Figs. 1(a) and 1(b)]. To determine the sign of J the sign of S_z should be known. The same experiment[6] suggests that the sign of S_z in the region of compensation is opposite to the equilibrium z-component sign of S_{Tz}, which is negative. Hence $S_z > 0$ and one obtains immediately $J > 0$. Actually the constant J entering Eq. (4) is determined by the *anisotropy* of the exchange interaction of pseudospins, $H_{\text{MFA}} = -\frac{J_{\|}}{2}\hat{\sigma}_z S_z - \frac{J_{\perp}}{2}(\hat{\sigma}_x S_x + \hat{\sigma}_y S_y)$,[15,16] which more generally includes two constants $J_{\perp} \neq J_{\|}$.[21] Taking into account that the terms like $\hat{\vec{\sigma}} \cdot \vec{S}$ in H_{MFA} do not affect the precessional term in Eq. (3) we conclude that the effective exchange constant in Eq. (4) for the Larmor frequency is $J = J_{\|} - J_{\perp}$. Constant $J_{\|}$ originates mainly from the exchange interaction of the carriers of the same name but belonging to different excitons,[22,23] whereas $J_{\perp}$ comes from the exciton-exciton exchange.[23] Also the interaction of polaritons with dark excitons and electron-hole plasma (under nonresonant excitation) may contribute to the constant J. Generally there are no fundamental limitations as to the sign of J.[22]

The initial pseudospin $\vec{S}^0$ is determined by two causes: (i) polarization of the incident light,[5] with the $\vec{S}^0$ components being related to the Stokes parameters P_l^0, $P_{l'}^0$, and P_c^0 of

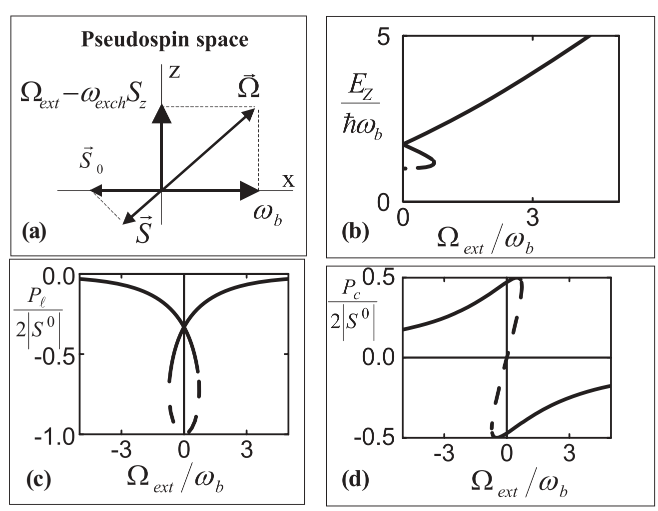

FIG. 2. (a) Orientation of the pseudospin vector under stationary conditions when the initial spin $\vec{S}^0$ is opposite to the x axis. (b) The enhanced Zeeman splitting of the levels $\pm 1/2$ vs Ω_{ext} frequency. The dependences of the degrees of linear (c) and circular (d) polarization of the polariton luminescence on the Ω_{ext} frequency. Solid (dashed) lines show stable (unstable) states.

the exciting light, as in Eq. (1); (ii) the anisotropy of the optical properties of the nanostructure, leading to the magnetic-field-independent linear dichroism effect and the anisotropy of recombination.[9] Interestingly, in the latter case the "initial" pseudospin $\vec{S}^0$ can be accumulated in the process of thermalization due to the polarization-selective escape of the cavity. In case (i) the $\vec{S}^0$ can be manipulated through the laser polarization.[5,9–12] In contrast to that, in case (ii) the vector $\vec{S}^0$ is fixed by the specific structural geometry. Nevertheless, one can change the orientation of $\vec{S}^0$ via externally applied uniaxial stress. The reversal of the $\vec{S}^0$ direction converts the suppression of Zeeman splitting into its enhancement. Even the spontaneous circular polarization of polaritons may take place in a nonequilibrium condensate. Indeed, let the initial pseudospin of polaritons $\vec{S}^0$ be directed opposite to the x axis. As is seen from Fig. 2(a), in this case the projection S_z onto $\Omega_{\rm ext}$ is negative, and the effective exchange field will be added to the external one. It will result in the enhancement of Zeeman splitting rather than suppression. It can be said that a negative feedback converts to a positive. This leads to the multivalued dependence $\vec{S}(\Omega_{\rm ext})$ in Fig. 2 (the parameter $a = -3$), with the stability of stationary states being determined with the help of the dynamic equation (3). Stable (unstable) states are shown by solid (dashed) lines in Fig. 2. If the exchange constant is sufficiently large ($\omega_{\rm exch}|S^0| > \omega_b$), a spontaneous circular polarization $S_z \neq 0$ will appear [Fig. 2(d)], sustained by the exchange field.[24] Such a state will be characterized by an appreciable Zeeman splitting in the exchange field even at $\Omega_{\rm ext} = 0$ [Fig. 2(b)].

VI. CONCLUSION

Thus, a simple model has been analyzed which describes the steady-state regime of a spin nonequilibrium homogeneous Bose-Einstein condensate of exciton polaritons. It explains all basic experimental results on the condensate polarization as well as the suppression of Zeeman splitting together with the unusual sign of the circular polarization. In turn, it predicts inverse effects, to wit, the spontaneous circular polarization in a zero external magnetic field and the enhancement of Zeeman splitting of a nonequilibrium condensate.

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