Stochastic polarization formation in exciton-polariton Bose-Einstein condensates

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We demonstrate theoretically the spontaneous formation of a stochastic polarization in exciton-polariton Bose-Einstein condensates in planar microcavities under pulsed excitation. Below the threshold pumping intensity (dependent on the polariton lifetime), the average polarization degree is close to zero, while above threshold the condensate acquires a polarization described by a (pseudospin) vector with random orientation, in general. It is shown that the polariton-polariton interaction leads to suppression of the linear polarization degree of the condensate due to the self-induced Larmor precession of the pseudospin. We establish the link between the second-order coherence of the polariton condensate and the distribution function of its polarization. We examine also the mechanisms of polarization dephasing and relaxation.

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I. INTRODUCTION

Bose-Einstein condensation (BEC) of exciton polaritons in semiconductor microcavities has recently been demonstrated experimentally.[1–3] It is still important, however, to establish a reliable and easily accessible experimental criterion for the BEC of polaritons. From the point of view of the Landau theory of phase transitions,[4] BEC requires the build up of an order parameter physically associated with the macroscopic wave function of the polariton condensate.[5] The phase of the order parameter is chosen by the system undergoing BEC spontaneously and this spontaneous symmetry breaking[6] is considered the "smoking gun" for BEC. The order-parameter buildup is also accompanied by a decrease in the second-order coherence parameter $g^{(2)}(0)$. Unfortunately, due to the limited time resolution of the Hanbury-Brown and Twiss experimental setup, it is very hard to measure $g^{(2)}(0)$ with a good accuracy.[7,8]

In recent works,[9–11] it has been suggested that the buildup of the order parameter can be evidenced by polarization measurements. Effectively, the polarization degree of light emitted by polariton condensates contains information on the amplitudes and relative phases of both components of the spinor wave function of the condensate. In this paper, we present a kinetic model of spontaneous formation of the polarization vector, which accompanies BEC of exciton polaritons in microcavities under pulsed excitation. We find a correlation between the polarization degree of light emitted by the microcavity and the second-order coherence of the polariton condensate and study depolarization of the condensate due to polariton-polariton interactions.

There have been a number of theoretical works describing the dynamics of polariton BEC. The approaches most relevant to our problem have been based either on the semiclassical Boltzmann equations describing the energy relaxation of polaritons but neglecting the phase of the condensate[12,13] or on the Gross-Pitaevskii equation and its generalizations, assuming the existence of a coherent condensate from the very beginning.[14–16] The spin dynamics of exciton polaritons has been treated within these two approaches as well.[10,17] The stochastic classical field model for polariton BEC was recently presented by Wouters and Savona.[18] Their "truncated Wigner" approach allowed calculating the second-order coherence of the condensate and recovering the Boltzmann equations in the low-density limit. The spin of exciton polaritons has been neglected however. In the recent work of del Valle *et al.*,[19] the buildup of linear polarization in a polariton BEC has been studied theoretically and experimentally in the presence of pinning. Our present model bridges the gap between the spin-dependent Boltzmann and spin-dependent Gross-Pitaevskii equations and allows one to describe the formation of a coherent polariton condensate from an incoherent ensemble of polaritons fully accounting for the polariton spin.

The formation of a polariton condensate is stochastic in its nature since polaritons entering the condensate have random phases and polarizations. Above the stimulation threshold, when the average population of the lowest-energy polariton state exceeds 1, both the stochastic phase and polarization of the condensate start being amplified and stabilize due to the stimulated scattering of polaritons from an incoherent reservoir. Polariton interactions with acoustic phonons and between themselves make the dynamics of the order-parameter complex and nontrivial since they lead to dephasing and polarization relaxation.

These general features of polariton condensate formation become especially noticeable under conditions of pulsed excitation, and we therefore focus on this regime in this work. We show that contrary to the case of cw excitation, where the polariton condensation is manifested by the spontaneous linear polarization formation,[1,2,9,10] the behavior of condensate polarization during the luminescence pulse is much more complex and interesting. This behavior is characterized by the interplay between the effects of polariton-polariton interactions, polarization relaxation, and polarization pinning. As a result, the condensate can exhibit suppression of linear polarization accompanied with the formation of rings in the circular component in the case when polarization relaxation is not effective. It is only the presence of effective and fast relaxation that leads to the suppression of circular polarization and to the formation of linear polarization as in the cw excitation case.

In this paper, we consider polariton condensation into one spin-degenerate-localized state. The generalization of the present theory to the case of several localized states and, in particular, the study of the spatial coherence formation that accompanies the polariton condensation will be given elsewhere. Neglecting the spatial degrees of freedom of the condensate in this paper is equivalent to the assumption of spatial coherence across the whole condensate. This is indeed the case in spatially confined systems, such as micropillars,[20] and is likely to be a feature of localized condensates in planar microcavities, as soon as the polariton lasing threshold is overcome.[11] We underline that while spatial coherence is an experimentally demonstrated feature[1] of BEC, it is not a sufficient condition or unambiguous signature. A simple argument is that spatial coherence has been observed in the microcavity optical parametric oscillator.[21–23] In this case, the spatial coherence is transferred from the laser excitation to the polariton field, but there is no spontaneous symmetry breaking or BEC.

For a localized condensate, the impact of the polariton-polariton interaction on the polarization dynamics depends on the localization radius: the larger the area occupied by the condensate the smaller the interaction effects for a fixed total number of polaritons. So, in reality one can have the case when polariton-polariton interaction effects strongly dominate the relaxation and dephasing effects (for strongly localized condensates) or the opposite case (for large localization radii). In Sec. II we present the theoretical formalism; in Sec. III we will present the results on the dynamics of polariton condensate pseudospin for the case when spin relaxation is negligible, but the polariton-polariton interactions are taken into account. Then in Sec. IV we consider the effects of fast relaxation. Finally, our conclusions are presented in Sec. V.

II. FORMALISM

The quantum kinetic equation for the condensate density matrix $\hat{\varrho}$ for the case of polariton condensation into one spatially localized state can be written as

$$\frac{d\hat{\varrho}}{dt} = \frac{i}{\hbar}[\hat{\varrho},\hat{H}] - \frac{1}{2}\sum_{\sigma=\pm 1}\{W(t)(\hat{a}_\sigma\hat{a}_\sigma^\dagger\hat{\varrho} + \hat{\varrho}\hat{a}_\sigma\hat{a}_\sigma^\dagger - 2\hat{a}_\sigma^\dagger\hat{\varrho}\hat{a}_\sigma) + \Gamma_c(\hat{a}_\sigma^\dagger\hat{a}_\sigma\hat{\varrho} + \hat{\varrho}\hat{a}_\sigma^\dagger\hat{a}_\sigma - 2\hat{a}_\sigma\hat{\varrho}\hat{a}_\sigma^\dagger)\}. \quad (1)$$

Here $\hat{a}_\sigma^\dagger$ and $\hat{a}_\sigma$ are the creation and annihilation operators of polaritons with the pseudospin projection $\sigma = \pm 1$, where the plus (minus) sign corresponds to the right (left) circular polarization.

The first term in the right-hand side of Eq. (1) describes the coherent evolution of the density matrix under the action of the condensate Hamiltonian $\hat{H}$. In what follows, we neglect the processes of virtual excitation of polaritons to the other orbital states, which can occur due to the polariton-polariton interaction, assuming the excited states to be far away in energy compared to the value of on-site interaction $\hat{V}$. This approximation is valid, in particular, for localized condensates in planar microcavities and for condensates in pillar microcavities. The condensate Hamiltonian can be written as

$$\hat{H} = \hat{H}_0 + \hat{V}, \quad (2a)$$

$$\hat{H}_0 = -\frac{1}{2}\hbar\Omega\sum_{\sigma\sigma'} C_{\sigma\sigma'}\hat{a}_\sigma^\dagger\hat{a}_{\sigma'}, \quad (2b)$$

$$\hat{V} = \frac{1}{2}\sum_{\sigma=\pm 1}\{\alpha_1\hat{a}_\sigma^\dagger\hat{a}_\sigma^\dagger\hat{a}_\sigma\hat{a}_\sigma + \alpha_2\hat{a}_\sigma^\dagger\hat{a}_{-\sigma}^\dagger\hat{a}_\sigma\hat{a}_{-\sigma}\}. \quad (2c)$$

Here the first term $\hat{H}_0$ describes possible polarization splitting of the single-polariton states, with Ω being the splitting frequency. The splitting matrix $C_{\sigma\sigma'}$ can be written as

$$C = \begin{pmatrix} c_z & c_x - ic_y \\ c_x + ic_y & -c_z \end{pmatrix}, \quad (3)$$

so that the matrix elements of C define the unit three-dimensional (3D) vector $\mathbf{c}$. For example, if the splitting of the condensate is because of an external magnetic field applied along z direction, $c_z=1$ and $c_x=c_y=0$. If there is no Zeeman splitting, $c_z=0$ and the vector $\mathbf{c}$ defines the linear polarization splitting of the condensate in the xy plane, which may be caused by the polarization splitting of exciton or photon modes forming the exciton polariton.

The polariton-polariton interaction $\hat{V}$ is characterized by two constants $\alpha_{1,2}$. It should be noted that these interaction constants are inversely proportional to the localization area of the condensate A. The localization area is found from the orbital wave function $\Phi(\mathbf{r})$ of the condensate according to the relation

$$\frac{1}{A} = \int d^2r|\Phi(\mathbf{r})|^4. \quad (4)$$

The interaction constant for polaritons with the same pseudospin σ can be estimated as $\alpha_1 \sim E_b a_B^2/A$, where a_B is the exciton Bohr radius and E_b is the exciton binding energy.[24] One expects a weak attraction between the polaritons with opposite pseudospins,[25] so that $\alpha_2 < 0$. The exact value of this parameter depends strongly on the number of quantum wells in the microcavity, their separation, and the detuning between the exciton and photon frequencies that define the exchange scattering of two polaritons.

The two other terms in Eq. (1) describe processes of incoherent income of polaritons into the condensate and the processes of polariton escape from the condensate. The income rate $W(t)$ is time dependent for the case of pulsed excitation and its value will be defined later in this section. The outcome rate of polaritons is mainly related to the finite transparency of the distributed Bragg mirrors of the microcavity and, therefore, can be described by a time-independent constant $\Gamma_c = 1/\tau_c$, where τ_c is the lifetime of polaritons in the condensate (typically a few picoseconds).

Quantum kinetic equations of type (1) for spinless polaritons have been studied previously for the cw excitation case, i.e., for the time-independent income rate W (see Ref. 26) and taking into account the time dependence of W but neglecting polariton-polariton interactions.[27,28] When both the interactions and the time dependence of rates are present, an analytical treatment of Eq. (1) is not feasible. To analyze the kinetics of polariton condensation numerically, we first transform Eq. (1) into the partial differential equation of the Fokker-Planck type. This is achieved by the Glauber-Sudarshan representation of the condensate density matrix[29]

$$\hat{\varrho} = \int d^4\psi \mathcal{P}(\psi,\psi^*)|\psi\rangle\langle\psi|. \tag{5}$$

Here we denote by ψ two complex numbers $\psi \equiv \{\psi_{+1}, \psi_{-1}\}$, so that the integration is over two complex planes $d^4\psi \equiv d^2\psi_{+1} d^2\psi_{-1}$, and the coherent states $|\psi\rangle$ are defined as

$$|\psi\rangle = \prod_{\sigma=\pm 1} \exp\{\psi_\sigma \hat{a}_\sigma^\dagger - \psi_\sigma^* \hat{a}_\sigma\}|\text{vac}\rangle. \tag{6}$$

After substitution of Eq. (5) into Eq. (1), making use of the relations

$$\hat{a}_\sigma|\psi\rangle\langle\psi| = \psi_\sigma|\psi\rangle\langle\psi|, \tag{7a}$$

$$\hat{a}_\sigma^\dagger|\psi\rangle\langle\psi| = \left(\psi_\sigma^* + \frac{\partial}{\partial\psi_\sigma}\right)|\psi\rangle\langle\psi|, \tag{7b}$$

and integration by parts we obtain the equation for the distribution function $\mathcal{P}(\psi,\psi^*)$. This equation reads as

$$\begin{aligned}\frac{\partial\mathcal{P}}{\partial t} = \sum_\sigma \Bigg\{ & -\frac{1}{2}\left[\sum_{\sigma'} F_{\sigma\sigma'}(t)\frac{\partial(\psi_{\sigma'}\mathcal{P})}{\partial\psi_\sigma} + \text{c.c.}\right] + \frac{W(t)}{2}\frac{\partial^2\mathcal{P}}{\partial\psi_\sigma\,\partial\psi_\sigma^*} \\ & + \frac{i}{\hbar}\left[\alpha_1\frac{\partial(|\psi_\sigma|^2\psi_\sigma\mathcal{P})}{\partial\psi_\sigma} + \alpha_2\frac{\partial(|\psi_{-\sigma}|^2\psi_\sigma\mathcal{P})}{\partial\psi_\sigma} - \text{c.c.}\right] \\ & - \frac{i}{2\hbar}\left[\alpha_1\frac{\partial^2(\psi_\sigma^2\mathcal{P})}{\partial\psi_\sigma^2} + \alpha_2\frac{\partial^2(\psi_\sigma\psi_{-\sigma}\mathcal{P})}{\partial\psi_\sigma\,\partial\psi_{-\sigma}} - \text{c.c.}\right]\Bigg\},\end{aligned} \tag{8}$$

where

$$F_{\sigma\sigma'}(t) = [W(t) - \Gamma_c]\delta_{\sigma\sigma'} + i\Omega C_{\sigma\sigma'}. \tag{9}$$

The first three terms in Eq. (8) are of the usual Fokker-Planck type. The first and the third terms containing the first derivatives are the so-called drift terms, while the second term describes the diffusion in the order-parameter ψ space. The fourth term in Eq. (8) is anomalous. In spite of its similarity to the diffusion term, it is characterized by negative diffusion coefficients. This term describes pure quantum kinetics that does not have a classical analog.

For polariton condensates in semiconductor microcavities, the polariton-polariton interaction starts to play a role only for sufficiently large condensate occupation, namely, in the region $\alpha_{1,2}\langle n\rangle \gtrsim \Gamma_c$. This implies a large average occupation of the condensate $\langle n\rangle \gg 1$, for typical values of $|\alpha_{1,2}| \lesssim 0.1$ meV and $\Gamma_c \sim 1$ ps^{-1}. For large occupations, the kinetics induced by the polariton-polariton interaction becomes semiclassical and the last term in Eq. (8) can be neglected. This term is small compared to the third one by the factor of $\langle n\rangle^{-1}$. In what follows, we will assume this limit and will omit the last term in Eq. (8). We note that neglecting this term is equivalent to the Gross-Pitaevskii approximation.

In the case of pulsed excitation, the initial condition for Eq. (8) is $\mathcal{P} = \delta^4(\psi)$ before the arrival of the excitation pulse, which implies the absence of polaritons in the condensate. The solution to Eq. (8) then gives the distribution function to calculate the statistical averages over many pulses. During each pulse, the evolution of the order parameter $\psi_\sigma(t)$ is random and can be described by a stochastic Langevin-type equation. The Langevin stochastic approach will be more convenient for us, and the Langevin equation that is equivalent[30] to Eq. (8) with the omitted last term is

$$\frac{d\psi_\sigma}{dt} = \frac{1}{2}[W(t) - \Gamma_c]\psi_\sigma + \theta_\sigma(t) - \frac{\mathrm{i}}{\hbar}\frac{\delta\mathcal{H}}{\delta\psi_\sigma^*}. \tag{10}$$

Here the first term describes evolution of the order parameter due to the pump and decay, the second term is the noise defined below that leads to the diffusive evolution of ψ_σ^*, and, finally, the third term appears due to the combined effect of the ground-state splitting and polariton-polariton interaction. This last term in the right-hand side of Eq. (10) has the same form as in the Gross-Pitaevskii equation, and we have written it as the functional derivative of the effective Hamiltonian function $\mathcal{H}$ of the order parameter. This function can be found from the condensate Hamiltonian (1) by replacing the creation and annihilation operators $\hat{a}_\sigma^\dagger$ and $\hat{a}_\sigma$ with ψ_σ^* and ψ_σ, respectively,

$$\mathcal{H} = -\frac{1}{2}\hbar\Omega\sum_{\sigma\sigma'} C_{\sigma\sigma'}\psi_\sigma^*\psi_{\sigma'} + \frac{1}{2}\sum_\sigma [\alpha_1|\psi_\sigma|^4 + \alpha_2|\psi_\sigma|^2|\psi_{-\sigma}|^2]. \tag{11}$$

The total intensity of the white complex noise $\theta_\sigma(t)$ is given by the income rate of polaritons into the condensate $W(t)$ that plays the role of the diffusion coefficient in the Fokker-Planck equation. The correlators of the noise are

$$\langle\theta_\sigma(t)\theta_{\sigma'}(t')\rangle = 0, \tag{12a}$$

$$\langle\theta_\sigma(t)\theta_{\sigma'}^*(t')\rangle = \frac{1}{2}W(t)\delta_{\sigma\sigma'}\delta(t-t'). \tag{12b}$$

This noise is responsible for the phase and polarization fluctuations in the ground state of the polariton system both below and above the condensation threshold.

It should be noted that the amplitude of the noise depends only on the value of income rate $W(t)$. The outcome rate of polaritons from the condensate given in our case by Γ_c does not affect the magnitude of the noise. Physically, it happens because the escape of particles from the condensate does not change the condensate coherence.[31] Mathematically, the noise term in the Langevin equation (10) appears from the diffusion (second) term in the Fokker-Planck equation (8). The easiest way to relate these terms is to consider the case of low condensate occupations ($|\psi_\sigma| \ll 1$), where only the diffusion term and the noise term can be kept in the Fokker-Planck and the Langevin equations, respectively. The amplitude of the noise can be then found by the comparison of the time dependence of the average number of polaritons.

In general, the income rate $W(t)$ should be found by solving the semiclassical Boltzmann equation for the polariton relaxation into the condensate.[12] In what follows, however, we adopt a simple model[15] considering all the polaritons that are not in the condensate as a single incoherent reservoir. The reservoir occupation number $N_r(t)$ satisfies the kinetic equation

$$\frac{dN_r}{dt} = -\Gamma_r N_r - W(t)[n(t)+1] + P(t), \tag{13}$$

where $P(t)$ is the incoherent pump rate, Γ_r^{-1} is the lifetime of polaritons in the reservoir (usually $\Gamma_r \ll \Gamma_c$), and $n(t) = |\psi_{+1}(t)|^2 + |\psi_{-1}(t)|^2$ is the instantaneous condensate occupation. The exact dependence of the income rate $W(t)$ on N_r is defined by the relaxation mechanism. In the simplest case of polariton-phonon relaxation, they are proportional to each other, $W(t) = rN_r(t)$.

In the case of very short pulsed excitation, with pulse duration much less then Γ_c^{-1}, the pump is reduced to the initial condition $N_r(0) = \int P(t)dt$ for the reservoir concentration. Equation (13) with $P=0$ is then solved simultaneously with Eq. (10) considering that the condensate is not initially populated, i.e., $\psi_\sigma(t=0)=0$. These equations were solved numerically using a (fifth-order) Adams-Bashforth-Moulton predictor-corrector method.[32] The results are presented in the next section.

III. FORMATION AND DYNAMICS OF THE ORDER PARAMETER

The complex order parameter of the condensate $\psi_\sigma(t)$ cannot be observed directly in a photoluminescence (PL) experiment. On the other hand, the polarization resolved PL gives access to the components of the condensate pseudospin (Stokes) vector,

$$S_x = (1/2)(\psi_{-1}^* \psi_{+1} + \psi_{+1}^* \psi_{-1}), \tag{14a}$$

$$S_y = (\mathrm{i}/2)(\psi_{-1}^* \psi_{+1} - \psi_{+1}^* \psi_{-1}), \tag{14b}$$

$$S_z = (1/2)(|\psi_{+1}|^2 - |\psi_{-1}|^2). \tag{14c}$$

Note that the condensate occupation n can be calculated from

$$n^2 = 4(S_x^2 + S_y^2 + S_z^2). \tag{15}$$

Averaged values and statistics of different experimentally observable quantities are presented in this section. In what follows, we will be interested mostly in time-integrated quantities, which will be denoted by a bar, e.g., $\int \psi_\sigma dt = \overline{\psi_\sigma}$. The averaging over multiple pulses (i.e., over realizations of noise) will be denoted by angular brackets as in Eq. (11).

From the definition, the time-averaged value of the pseudospin $\overline{\mathbf{S}}$ provides direct information about the order parameter of the condensate (except for the phase). It should be noted that fluctuations in the direction of the vector $\overline{\mathbf{S}}$ appear due to the same physical reasons as fluctuations of the condensate phase, namely, due to the income of polaritons into the condensate with random polarizations and phases. Therefore, suppression of fluctuations of $\overline{\mathbf{S}}$ indicates suppression of fluctuations of the phase of the condensate as well and manifests in the buildup of the order parameter.

When describing experiments on polariton Bose-Einstein condensation in microcavities, one may be able to neglect the effects of polariton-polariton interactions and/or polarization splitting of the ground state of the condensate depending on the sample, geometry of the experiment, and pumping intensity. The polarization splitting typically depends on the position of the excitation spot and the polariton-polariton interaction decreases with the increase in the condensate localization radius (for constant occupation number of the condensate). For this reason, in what follows, we present first the results obtained neglecting both these effects [i.e., setting $\mathcal{H}=0$ in Eq. (10)], then we include the effect of polariton-polariton interactions, and finally we study the role of polarization splitting. In numerical calculations, we used the parameters $\Gamma_c = 0.5\ \text{ps}^{-1}$, $\Gamma_r/\Gamma_c = 0.01$, $r = 10^{-4}\ \text{ps}^{-1}$, and, when the polariton-polariton interaction is taken into account, $\alpha_1 = 1.8\ \mu\text{eV}$ and $\alpha_2/\alpha_1 = -0.1$ (see Ref. 33).

A. Noninteracting BEC without polarization splitting

To evidence the order-parameter formation, it is convenient to study the total polarization degree of the condensate (TPDC) defined as

$$\rho = \frac{2}{\overline{n}}[(\overline{S_x})^2 + (\overline{S_y})^2 + (\overline{S_z})^2]^{1/2}. \tag{16}$$

The TPDC changes from 0 for a chaotic state to 1 for the case of a well-defined order parameter with suppressed fluctuations in time. It can be calculated numerically for different realizations of the noise as Fig. 1 shows. One can see that the solutions to Eqs. (10) and (13) exhibit a well-pronounced threshold behavior. The dynamical threshold[27] is defined by the balance of the polariton income and outcome rates for the condensate $W(0) = rN_r(0) = \Gamma_c$, so that the threshold pump intensity is given by $\int P_{th} dt = \Gamma_c / r$.

Below threshold, the average occupation of the condensate is less than unity (see Fig. 2) and the order parameter fluctuates extensively during the reservoir lifetime Γ_r^{-1}, which defines the duration of the PL signal in this case. The pseudospin also fluctuates strongly both in amplitude and direction, so that the TPDC is close to zero. Because of the finite duration of the PL signal (also given by Γ_r^{-1}), the TPDC is not averaged to zero exactly, and it can be shown that $\langle \rho \rangle \sim (\Gamma_r/\Gamma_c)^{1/2}$ for $\Gamma_r \ll \Gamma_c$ and $P \ll P_{th}$.

Above threshold, the condensate is formed during the formation time t_f, such that $W(t) \geq \Gamma_c$ for $0 < t \leq t_f$, and disappears afterward on the scale of Γ_c^{-1}. This leads to a drastic

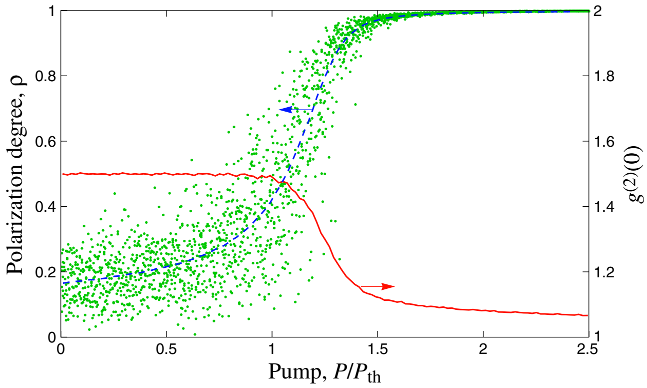

FIG. 1. (Color online) The total polarization degree ρ of a polariton condensate as a function of pump intensity for the case of noninteracting polaritons. Dots show the random values of ρ for different pulses, while the average $\langle\rho\rangle$ is shown by the dashed line. The solid line shows the second-order coherence parameter (see text).

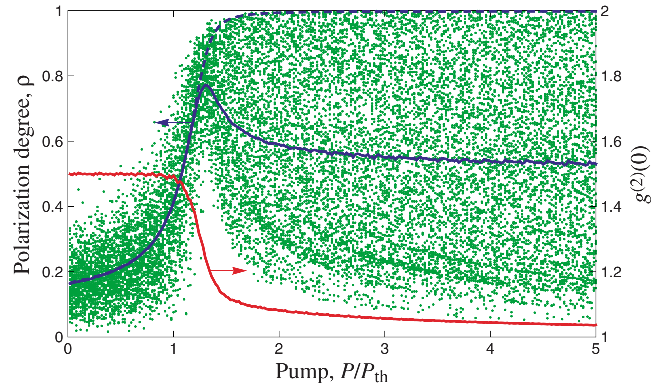

FIG. 3. (Color online) The same as Fig. 1 but accounting for polariton-polariton interactions. Dots show again the random values of ρ for different pulses, while the average $\langle\rho\rangle$ is shown by the solid (blue) line. It is seen that interactions decrease the total polarization degree with respect to the noninteracting case (dashed line). The other solid (red) line shows the second-order coherence parameter.

increase in the condensate occupancy and narrowing of the emission peak (see Fig. 2). At the same time, there is a strong increase in the total polarization degree of the condensate. The TPDC reaches unity, as the dashed curve in Fig. 1 shows. The fluctuations of the order parameter become fully suppressed for $P \gg P_{th}$, so that apparently the fluctuations of ρ are most pronounced in the vicinity of the threshold. It should be noted that while the buildup of TPDC indicates the formation of an order parameter for each excitation pulse, the values of the order parameter and the corresponding values of pseudospin change randomly from pulse to pulse.

Formation of the coherent polariton state at $P > P_{th}$ can be observed also by measuring the second-order coherence in a Hanbury-Brown and Twiss setup.[7,8] The second-order coherence parameter $g^{(2)}(0)$,

$$g^{(2)}(0) = \frac{\overline{\langle n^2\rangle}}{\overline{\langle n\rangle^2}}, \tag{17}$$

is also shown in Fig. 1. Note that in contrast to the TPDC, $g^{(2)}(0)$ is not a direct measure of the order parameter, while its power dependence is sensitive to the buildup of the order parameter. The increase in the pumping power brings the polariton condensate from the thermal (chaotic) state with $g^{(2)}(0) = 3/2$ for $P \ll P_{th}$ (Ref. 34) to the coherent state for $P \gg P_{th}$, where the fluctuations are suppressed and the order parameter is well defined for each pulse.

B. Effects of polariton-polariton interactions

Strong polariton-polariton interactions modify drastically the dependence of the TPDC on the pump, as it is shown in Fig. 3. They induce a substantial increase in fluctuations of the polarization degree. Another important effect of polariton-polariton interactions is the nonuniform distribution of the total polarization degree for $P > P_{th}$. Apart from strong fluctuations of the polarization degree from 0 to 1, its distribution exhibits sharp and approximately equidistant peaks that appear far above threshold, as shown in Fig. 4. The origin of these peaks can be understood if one examines the condensate polariton distribution function in pseudospin space. This distribution function is shown in Fig. 5 for four values of the pump power.

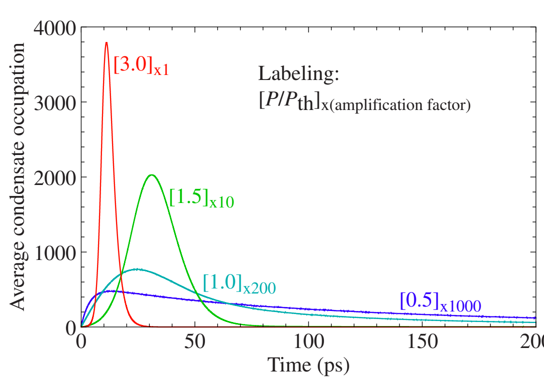

FIG. 2. (Color online) The time dependence of the average condensate occupation number $\langle n\rangle$ is shown for the same values of P/P_{th}. The curves are scaled according to the subindex in the labels.

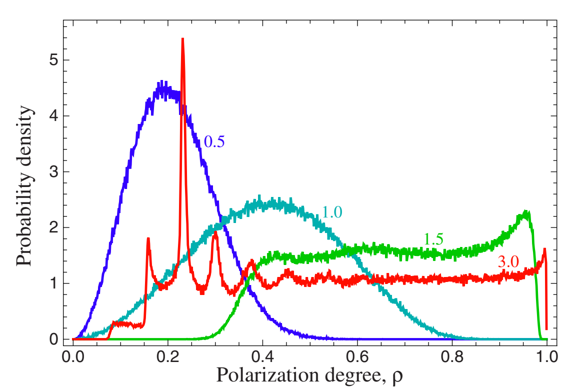

FIG. 4. (Color online) The distribution function of the total polarization degree shown in Fig. 3. Curves are labeled by the values of P/P_{th}.

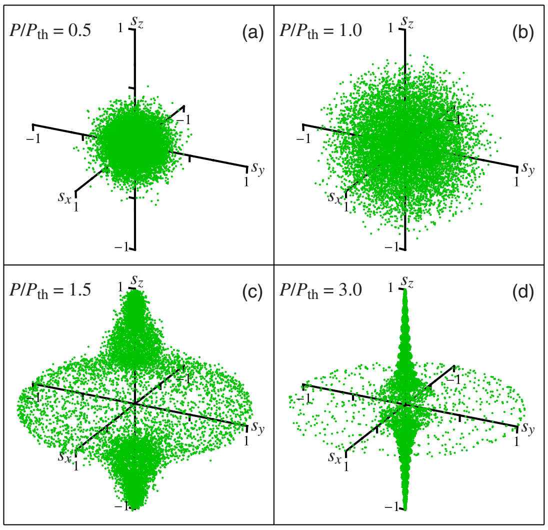

FIG. 5. (Color online) The distribution function in normalized pseudospin space $\mathbf{s}=2\overline{\mathbf{S}}/\bar{n}$ for different values of the excitation pump in the case of interacting polaritons. Above the threshold, the formation of the Larmor rings is seen.

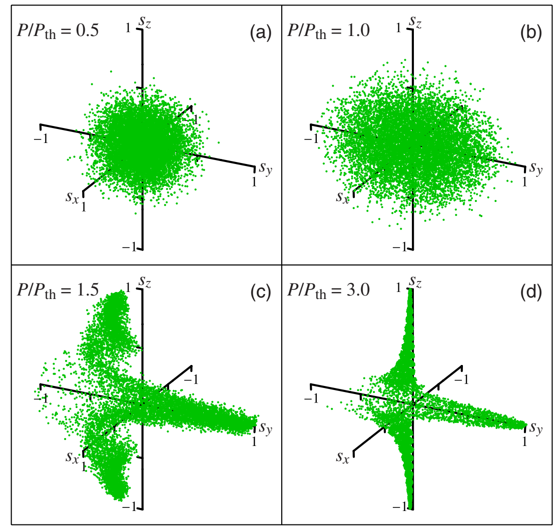

FIG. 6. (Color online) The distribution function in normalized pseudospin space $\mathbf{s}=2\overline{\mathbf{S}}/\bar{n}$ in the case of polarization splitting characterized by $\mathbf{c}\parallel\hat{y}$ with the splitting energy $\hbar\Omega=0.08$ meV.

One can see that the in-plane component of pseudospin $(S_x^2+S_y^2)^{1/2}$ is strongly suppressed for several values of S_z [see Fig. 5(d) for $P=3P_{th}$]. This happens as a result of the spin anisotropy of polariton-polariton interactions. This anisotropy produces the self-induced Larmor precession[17] of the pseudospin vector around the $\hat{z}$ axis. The frequency of this precession is proportional to S_z (see the Appendix for more details). For a fixed n, the in-plane component of pseudospin is averaged to zero for the values of S_z that give a full 2π rotation. In reality, the condensate occupation is not fixed and n also fluctuates, so that the in-plane pseudospin never averages to zero exactly. Nevertheless, this effect gives rise to a sequence of "Larmor rings" seen at strong pump intensity. An important consequence of the suppression of the average in-plane component of the pseudospin is the nonmonotonous dependence of the TPDC on the pump intensity. The TPDC exhibits a peak placed close to the threshold value of the pump. The nonmonotonic behavior of the TPDC as a function of pump intensity with a characteristic peak near the threshold pump intensity has been observed experimentally in GaN-based microcavities.[11]

Note that the polariton-polariton interactions that caused a decrease in the average polarization degree for higher pump powers do not prevent $g^{(2)}$ from approaching 1, and the interactions have no effect on the second-order coherence (cf. Figs. 1 and 3). This is because of the definition of the second-order coherence parameter at an instantaneous value of time [see Eq. (17)]. Experimentally, $g^{(2)}(0)$ is measured by a Hanbury-Brown and Twiss setup, which has a temporal resolution of about 100 ps, typically. Both below and much above the stimulation threshold, this value exceeds the first-order coherence time of the condensate,[8] which is why the measured value of $g^{(2)}(0)$ always remains close to zero. When corrected accounting for the nonmonotonic dependence of the first-order coherence time on the pumping power $g^{(2)}(0)$ exhibits the decrease above threshold[7] followed in a CdTe microcavity by subsequent increase with the pumping power.[8] The decrease of $g^{(2)}(0)$ with the pumping power is reproduced by the Wigner model of Wouters and Savona[18] and by our previous[28] and present model. The nonmonotonic behavior above threshold could reflect the multimode character of emission of the cavity and fluctuations of the condensate energy due to fluctuations of its occupation number $n(t)$ within the time resolution of the experimental setup. The effect of fluctuations of the condensate occupation on the second-order coherence above threshold has been studied in detail experimentally and theoretically in Ref. 35. This effect can be described by our theory as well but is beyond the scope of this paper.

C. Combined effects of polarization splitting and interactions

The presence of polarization splitting described by the first term in the Hamiltonian (11) has a remarkable effect on the pseudospin distribution function, as shown in Fig. 6. The vector $\mathbf{c}$ that defines the splitting matrix (3) has been chosen along the $\hat{y}$ direction. One can see that the Larmor rings appearing at high pump become deformed and are accompanied by pinning of linear polarization to the $\hat{y}$ axis.

The presence of pinning seen in Fig. 6 is somewhat unexpected. Indeed, the pseudospin vector is initially formed with some random orientation. Then, in the absence of polariton-polariton interactions and relaxation, the pseudospin should simply precess around the $\hat{y}$ axis, and, since this precession conserves the y component of the pseudospin vector, the distribution function should remain symmetric, i.e., without any pinning. It turns out that pinning is a combined effect of polarization splitting and polariton interactions. The features of the precession in this case are presented in the Appendix. Physically, the appearance of pinning can be understood from energy conservation. Assume that initially the pseudospin vector is randomly oriented in the equatorial plane. For this orientation, the polariton-polariton repulsion is minimized. Polarization splitting will rotate the pseudospin vector and move it away from the equator. This results in an increase in the polariton repulsion energy and this increase should be compensated by a decrease in the splitting energy. Therefore, the angle between the vectors **c** and **S** will decrease, so that pinning appears.[36] It is also shown in the Appendix that for high enough polariton-polariton interaction, one still observes the self-induced Larmor precession along with the deformed precession around the pinning **c** direction. The axis of self-induced Larmor precession, however, becomes inclined to the direction opposite to the pinning axis. This explains the deformation of the Larmor rings in the $-\hat{y}$ direction, as it is seen in Fig. 6.

IV. RELAXATION OF THE ORDER PARAMETER

The results obtained in the previous section are valid if the lifetime of the condensate Γ_c^{-1}, which defines the duration of the luminescence signal far above the threshold, is short compared to the typical relaxation times of the order parameter. In this section, we show that fast relaxation of the order parameter modifies qualitatively the polarization properties of the polariton condensate.

The relaxation can be incorporated phenomenologically in our Eq. (10) by adding a relaxation term $\mathfrak{R}_\sigma$ to its right-hand side. In general, this should be accompanied by an additional noise term due to the fluctuation-dissipation theorem, but this additional noise is small compared to the income noise and can be neglected if the lattice temperature is not too high. In the simplest case, the so-called model A also referred to as the Landau-Khalatnikov or the Onsager model,[37] this relaxation term reads as

$$\mathfrak{R}_\sigma^{\mathrm{LK}} = -\nu\frac{\delta\mathcal{H}}{\delta\psi_\sigma^*}, \tag{18}$$

where the parameter ν defines the relaxation rate. This term favors relaxation of the order parameter to a state with $S_z=0$, i.e., to a linearly polarized condensate, since the polariton-polariton repulsion energy is minimized for the linear polarization.

The distribution functions of polaritons normalized in the pseudospin space calculated for different pump intensities in the absence of polarization splitting ($\Omega=0$) are shown in Fig. 7. One can see that the "Larmor rings" vanish and the circular polarization of the condensate is rapidly lost as the pump intensity increases. The distribution of polariton condensates in pseudospin space calculated by repeating the numerical experiment with different noise realizations changes from a sphere to a torus and approaches a flat ring shape as the pump power is increased further. As a result, the condensate develops a strong spontaneous linear polarization far above the condensation threshold. If the polarization splitting of the ground state is present, the direction of resulting linear polarization becomes well defined.

It should be noted that the relaxation term (18) does not conserve the number of polaritons in the condensate. This way the condensate depletion and leakage of polaritons from the condensate are implied by the model (18). In the case when the condensate occupation is not very high and only

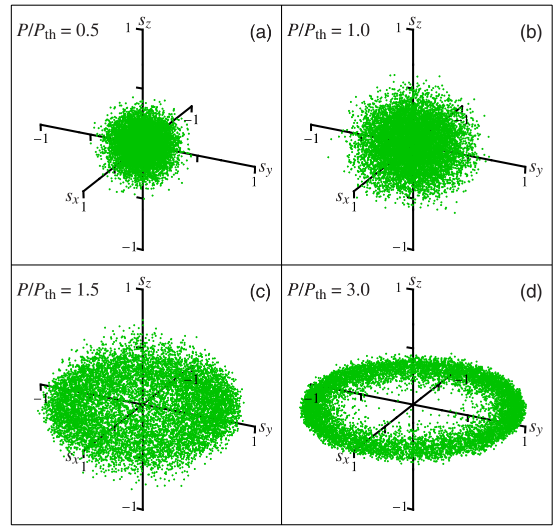

FIG. 7. (Color online) The effect of relaxation of the order parameter on the distribution function in normalized pseudospin space $\mathbf{s}=2\bar{\mathbf{S}}/\bar{n}$. Contrary to the case without relaxation (Fig. 5), a linear polarization is formed for strong pump above threshold. The relaxation parameter $\nu=1\ \mathrm{meV}^{-1}\ \mathrm{ps}^{-1}$.

the term linear in ψ_σ is kept in Eq. (18), the Landau-Khalatnikov relaxation is reduced to

$$\mathfrak{R}_\sigma^{\mathrm{lin}} = \frac{1}{2}\gamma\sum_{\sigma'} C_{\sigma\sigma'}\psi_{\sigma'}. \tag{19}$$

It is easily seen that this type of relaxation is equivalent to the assumption of an anisotropy in the transparency of the distributed Bragg mirrors of the microcavity, i.e., it is equivalent to the presence of polarization dependence of the polariton lifetime. In particular, the lifetime is $\tau_c=1/(\Gamma_c-\gamma)$ for polarization along **c**, and $\tau_c=1/(\Gamma_c+\gamma)$ for the perpendicular linear polarization. Clearly, this model makes sense only when the relaxation rate $\gamma<\Gamma_c$.

Probably, the simplest relaxation model that conserves the number of polaritons in the condensate is

$$\mathfrak{R}_\sigma^{\mathrm{cst}} = -\frac{1}{2}\gamma R_{\sigma\sigma'}\psi_{\sigma'}, \tag{20}$$

$$R_{\sigma\sigma'} = (\mathbf{c}\cdot\mathbf{S})\delta_{\sigma\sigma'} - SC_{\sigma\sigma'}. \tag{21}$$

Note that this type of relaxation is reflected in the term $-\gamma[\mathbf{S}\times[\mathbf{S}\times\mathbf{c}]]$ in the equation for the pseudospin velocity $d\mathbf{S}/dt$. If we denote θ as the angle between **S** and **c**, then this angle relaxes according to the equation $d\theta/dt=-\gamma S\sin\theta$. We will present elsewhere[38] the results for the case of occupation-conserving relaxation along with the results about junctions of several polariton BECs and formation of spatial coherence, where the conservation of the total number of particles plays a crucial role.

V. CONCLUSIONS

We have described theoretically the spontaneous buildup of the vector order parameter in the course of polariton BEC. The buildup of the order parameter manifests itself in the formation of the stochastic vector polarization of the condensate. This polarization is correlated with the second-order coherence of the condensate. We have considered the regime of pulsed excitation either neglecting or allowing for the polariton polarization (spin) relaxation in the condensate (Figs. 5 and 7, respectively). We also considered the effects of polarization splitting of the polariton state that results in linear polarization pinning. If the spin relaxation is inhibited, we predict a strong circular polarization of the condensate having an oscillating statistical distribution. If the spin relaxation is efficient, the condensate is expected to acquire a linear polarization.

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APPENDIX: PSEUDOSPIN DYNAMICS

Here we discuss the dynamics of the pseudospin vector **S** in the region of high excitation densities, i.e., for the pump far above the stimulation threshold.

It is necessary to point out that, in general, there is no closed equation for the pseudospin vector. Such an equation cannot be obtained from Eq. (10) and definitions (14a)–(14c) because of the noise term in Eq. (10) that cannot be transformed into a term depending solely on **S**. Physically, the absence of a closed equation for **S** is related to the fact that the pseudospin vector misses the information about the phase of the condensate and therefore does not provide the complete description of the system.

However, at high pump densities, i.e., far above the threshold, the relative fluctuations of the order parameter are small and the noise term can be omitted. Also, if we are interested in the dynamics of the order parameter on time scales where the condensate occupation does not change significantly, it is possible to neglect the (first) pump-decay term in Eq. (10) as well. Then the order parameter evolves according to the Gross-Pitaevskii equation

$$i\hbar\frac{d\psi_\sigma}{dt}=\frac{\delta\mathcal{H}}{\delta\psi_\sigma^*}=-\frac{1}{2}\hbar\Omega\sum_{\sigma'}C_{\sigma\sigma'}\psi_{\sigma'}+[\alpha_1|\psi_\sigma|^2\psi_\sigma+\alpha_2|\psi_{-\sigma}|^2\psi_\sigma]. \quad (A1)$$

This equation can be transformed to the equation for the pseudospin vector (14a)–(14c). Namely,

$$\frac{d\mathbf{S}}{dt}=\Omega[\mathbf{S}\times\mathbf{c}]+2\frac{(\alpha_1-\alpha_2)}{\hbar}(\hat{\mathbf{z}}\cdot\mathbf{S})[\hat{\mathbf{z}}\times\mathbf{S}], \quad (A2)$$

where we used the definition of the 3D vector **c** from Eq. (3).

For low condensate occupation, the nonlinear interaction term can be neglected and this equation describes the precession of the pseudospin vector **S** around the pinning direction

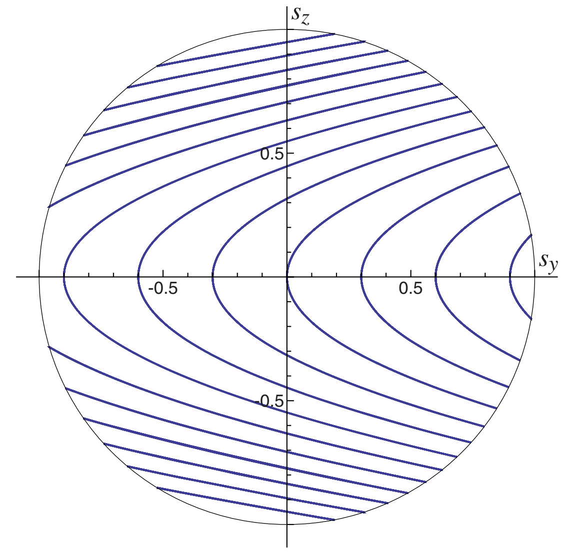

FIG. 8. (Color online) Showing the projections on the yz plane of several trajectories drawn by the end of the pseudospin vector on the Poincaré sphere for the case of combined precession discussed in the Appendix. The trajectories are defined by the parameter a ranging from −2.7 to 0.9 in increments of 0.3. The interaction parameter $\lambda=6$.

c. On the contrary, for large condensate occupation n the first term in Eq. (A2) can be neglected and the self-induced Larmor precession around the $\hat{z}$ axis appears due to the polariton-polariton interaction. The angular velocity of this precession depends on the value of S_z, which is the reason for the appearance of the Larmor rings discussed in Sec. III.

In the general case, Eq. (A2) also describes a periodic motion of the pseudospin. The value of $|\mathbf{S}|$ is conserved and the end of the vector **S** draws closed trajectories on the Poincaré sphere. To describe the experimentally relevant case of linearly polarized pinning, we choose the pining direction $\mathbf{c}\|\hat{\mathbf{y}}$, as in Sec. III C. Then, introducing the unit vector $\mathbf{s}=2\mathbf{S}/n$, we have

$$\frac{1}{\Omega}\frac{ds_x}{dt}=-s_z-\lambda s_z s_y, \quad (A3a)$$

$$\frac{1}{\Omega}\frac{ds_y}{dt}=\lambda s_z s_x, \quad (A3b)$$

$$\frac{1}{\Omega}\frac{ds_z}{dt}=s_x, \quad (A3c)$$

where

$$\lambda=\frac{(\alpha_1-\alpha_2)n}{\hbar\Omega}. \quad (A4)$$

The solutions to Eq. (A2) can be expressed in terms of Jacobi's elliptic functions,

$$s_z=A\,\mathrm{cn}(\omega t,k^2), \quad (A5a)$$

$$s_y=a+(1/2)\lambda s_z^2, \quad (A5b)$$

$$s_x = -\omega A \mathrm{sn}(\omega t, k^2)\mathrm{dn}(\omega t, k^2), \tag{A5c}$$

where the frequency ω, the elliptic modulus k, and the amplitude A can be found from

$$(\omega/\Omega)^4 = 1 + 2a\lambda + \lambda^2, \tag{A6}$$

$$A^2 = \frac{2(1-a^2)}{(1+a\lambda) + \sqrt{1+2a\lambda+\lambda^2}}, \tag{A7}$$

$$k^2 = \frac{1}{2}\left[1 - \frac{(1+a\lambda)}{\sqrt{1+2a\lambda+\lambda^2}}\right]. \tag{A8}$$

The number a in above equations is a free parameter that is defined by the initial condition. For the weak-interaction case, when $0<\lambda<1$, one has $-1<a<1$ and all the pseudospin trajectories cross the equatorial plane of the Poincaré sphere, so that one has a deformed precession around **c** axis. When $\lambda>1$, the parameter a can be smaller than -1, namely, $-(1+\lambda^2)/2\lambda<a<1$. The trajectories for $a<-1$ never reach the equator of the Poincaré sphere and they describe the precession analogous to the self-induced Larmor precession.[39] The precession axis, however, is inclined with respect to the $\hat{z}$ direction (see Fig. 8).

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