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 life-time) the average polarization degree is close to zero, whilst above threshold the condensate acquires a polarization described by a (pseudospin) vector with random orientation, in general. We establish the link between 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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Introduction.—Bose-Einstein condensation (BEC) of exciton-polaritons in semiconductor microcavities has recently been demonstrated experimentally [1,2,3]. This important discovery opens the perspective for various applications including room-temperature polariton lasers [4], microscopic optical parametric oscillators [5], and optical logic gates [6]. In this context, it is important to establish a reliable experimental criterion for the BEC of polaritons. From the point of view of the Landau theory of phase transitions [7], BEC requires the build up of an order parameter physically associated with the macroscopic wave-function of the polariton condensate [8]. The phase of the order parameter is chosen by the system undergoing BEC spontaneously; the spontaneous symmetry breaking [9] is considered as the “smoking gun” for BEC. The order parameter build up is also accompanied by a decrease of the second order coherence parameter, $g_2(0)$, indicating the change from a thermal distribution of polaritons to a coherent distribution [10]. Unfortunately, due to the limited time resolution of the Hanbury-Brown and Twiss experimental set up, it is very hard to measure $g_2(0)$ with a good accuracy [11,12].

In recent work [13,14] it has been suggested that the build-up 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 Letter we present the kinetic model of spontaneous formation of the polarization vector during the course of polariton BEC. Indeed, the polarization vector provides the same information on the condensate (complex) order parameter, as the second order coherence; this allows a straightforward method of evidencing the existence of spontaneous symmetry breaking in a polariton system under pulsed excitation.

Existing theories for the dynamics of polariton BEC have been based either on the semiclassical Boltzmann equations describing the energy relaxation of polaritons but neglecting the phase of the condensate [15], or on the Gross-Pitaevskii equation and its generalizations, assuming the existence of a coherent condensate from the very beginning [16,17,18]. The present model bridges the gap between these two approaches and allows one to describe the formation of a coherent polariton condensate from an incoherent ensemble of polaritons.

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, the stochastic phase and polarization of the condensate start being amplified and stabilized 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 non-trivial since they lead to dephasing and polarization relaxation. Here we treat these effects within the Landau-Khalatnikov approach [19] generalized to describe a two-component interacting Bose system.

Formalism.—We consider BEC into one spin-degenerate ground-state level. The evolution of the order parameter $\psi_\sigma(t)$ is described by the Langevin type equation:

$$\frac{d\psi_\sigma}{dt} = \frac{1}{2} [W(t) - \Gamma_c] \psi_\sigma + \theta_\sigma(t) - \frac{i}{\hbar} \frac{\delta H_{int}}{\delta \psi_\sigma} - \gamma R_\sigma. \quad (1)$$

Here $\sigma = \pm 1$ denotes the two spin (circularly polarized) components of the order parameter [20], $W(t)$ is the income rate, $\Gamma_c^{-1}$ is the polariton lifetime in the condensate, $\theta_\sigma(t)$ is the noise defined below. The third term in the righthand side of Eq. (1) describes the spin-dependent polariton-polariton interactions in the condensate

$$H_{int} = \frac{1}{2} [\alpha_1 (|\psi_{+1}|^4 + |\psi_{-1}|^4) + 2\alpha_2 |\psi_{+1}|^2 |\psi_{-1}|^2], \quad (2)$$
where $\alpha_1 > 0$ defines the repulsion energy of two polaritons with the same spin and $\alpha_2 < 0$ defines the attraction energy of two polaritons with opposite spins. Finally, the last term in Eq. (11) describes the relaxation of the order parameter. The form of the functional $R_{\sigma}\{\psi_{\sigma}, \psi_{\sigma}^*\}$ depends on the mechanism of relaxation.

In the particular case of low condensate occupations, when one can neglect the polariton-polariton interactions and the order parameter relaxation, Eq. (11) reduces to the kinetic equation for the order parameter derived in [21] and originated from the quantum kinetic equation describing the evolution of the condensate density matrix [22]. In this case the total intensity of the white noise, $\theta_{\sigma}(t)$, is given by the income rate of polaritons into the condensate, $W(t)$. The correlator of the noise is

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

(3)

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. Above threshold, when the condensate occupation $n(t) = |\psi_{+1}|^2 + |\psi_{-1}|^2$ is not small, dephasing due to polariton self-interaction [23] becomes also important.

In general, the income rate $W(t)$ should be found by solving the semiclassical Boltzmann equation for the polariton relaxation into the condensate [13]. In what follows, however, we adopt a simple model [17] considering all the polaritons that are not in condensate as a single incoherent reservoir. The reservoir occupation number $N_r(t)$ satisfies the kinetic equation:

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

(4)

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

In the case of very short pulsed excitation, with pulse duration much less then $\Gamma_c^{-1}$, the pump is reduced to the initial condition $N_r(0) = \int P(t) dt$ for the reservoir concentration. Eq. (4) with $P = 0$ is then solved simultaneously with Eq. (11) considering that the condensate is not initially populated, i.e., $\psi_{\sigma}(0) = 0$. First, we present the results for the case when the polarization relaxation can be neglected ($\gamma = 0$), and then we describe the effects of relaxation of the order parameter.

**Formation of the order parameter.**—The complex order parameter of the condensate $\psi_{\sigma}$ cannot be observed directly in a photoluminescence (PL) experiments. On the other hand, the polarization resolved PL gives access to the components of the condensate pseudospin (Stokes vector) [20]. These components are related to $\psi_{\sigma}$ as $S_x = (1/2)(\psi_{+1}^* \psi_{-1} + \psi_{+1} \psi_{-1}^*), S_y = (i/2)(\psi_{+1}^* \psi_{-1} - \psi_{+1} \psi_{-1}^*), S_z = (1/2)|\psi_{+1}|^2 - |\psi_{-1}|^2$. Note that the condensate occupation, $n$, can be calculated from $n^2 = 4(S_x^2 + S_y^2 + S_z^2)$. Averages and statistics of different experimentally observable quantities are presented in this subsection. In what follows we will be interested mostly in time-integrated quantities which will be denoted by a bar, e.g., $\bar{\bar{\psi}}_{\sigma} dt = \psi_{\sigma}$. The averaging over multiple pulses (i.e., over realizations of noise) will be denoted by angular brackets as in Eq. (3). In numerical calculations we used the parameters $\Gamma_c = 0.5$ ps$^{-1}$, $\Gamma_r/\Gamma_c = 0.01$, $\alpha_1 = 1.8$ meV, $\alpha_2/\alpha_1 = -0.1$, and $r = 10^{-4}$ ps$^{-1}$ [24].

The total polarization degree of the condensate,

$$\rho = \frac{2}{\pi} \left[ |\bar{\bar{S_x}}|^2 + |\bar{\bar{S_y}}|^2 + |\bar{\bar{S_z}}|^2 \right]^{1/2},$$

(5)

is shown in Fig. 1. One can see that the solutions to Eqs. (11) and (4) exhibit a well pronounced threshold behavior. The dynamical threshold [22] is defined by the balance of the polariton income and outcome rates for the condensate: $W(0) = r N_r(0) = \Gamma_c$, so that the threshold pump is defined by $\int P_{th} dt = \Gamma_c/\Gamma_r$. 

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**FIG. 1:** (a) The total polarization degree $\rho$ of polariton condensate as a function of pump intensity. Dots show the random values of $\rho$ for different pulses, while the average $\langle \rho \rangle$ is shown by the solid line. The other solid line shows the second-order coherence (see text). (b) The distribution function of the total polarization degree. Curves are labeled by the values of $P/P_{th}$. The time dependence of the average condensate occupation number $\langle n \rangle$ is shown in the insert for the same values of $P/P_{th}$.
Below threshold, the average occupation of the condensate is less than 1 and the order parameter fluctuates extensively during the reservoir lifetime $\Gamma_c^{-1}$ that defines the duration of the PL signal in this case. The pseudospin also fluctuates strongly both in amplitude and direction, so that the total polarization degree is close to zero. It can be shown that $\left\langle \rho \right\rangle \sim (\Gamma_c/\Gamma_r)^{1/2}$ 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 afterwards on the scale of $\Gamma_c^{-1}$. This leads to a drastic increase in the condensate occupation and narrowing of the emission [see insert in Fig. 1(b)]. At the same time there is a strong increase in the total polarization degree of the condensate. Without polariton-polariton interactions the polarization degree would reach 1. However, the polariton-polariton interactions [the third term in Eq. (1)] result in a slow decrease of the average polarization degree for $P > P_{th}$. As a result the dependence $\left\langle \rho \right\rangle$ on $P$ exhibits a maximum placed close to the threshold. The build-up of the total polarization degree indicates the formation of a well-defined order parameter and well-defined pseudospin for each excitation pulse. It should be noted, however, that the direction of the pseudospin randomly changes from pulse to pulse.

Formation of the order parameter at $P > P_{th}$ can be observed also by measuring the second-order coherence in Hanbury-Brown and Twiss experiment [11, 12, 25]. The second-order coherence parameter $g^{(2)}(0) = \left\langle n^2 \right\rangle / \left\langle n \right\rangle^2$ is also shown in Fig. 1(a). It is seen that increase of the pump brings the polariton condensate from the thermal (chaotic) distribution with $g^{(2)}(0) = 3/2$ for $P \ll P_{th}$ [26] to the coherent state with $g^{(2)}(0) \simeq 1$ for $P \gg P_{th}$, where the fluctuations are suppressed and the order parameter is well-defined for each pulse. Note that the polariton-polariton interactions that caused a decrease in the average polarization degree for higher pump powers do not prevent $g^{(2)}(0)$ from approaching 1. In other words the polariton-polariton interactions do not result in loss of coherence; rather they have a dynamical effect on the pseudospin vector, which affects the average polarization degree as we shall discuss later. Clearly, both the second-order coherence and polarization measurements provide evidences of the condensate formation. However, the polarization measurements give more details about the state of the condensate since the total polarization is sensitive to the effects of polariton-polariton interactions.

Another important effect of polariton-polariton interaction is the nonuniform distribution of the total polarization degree for $P > P_{th}$. Apart from strong fluctuations from 0 to 1 present in this case the distribution of polarization degree exhibits sharp and approximately equidistant peaks that appear far above threshold as shown in Fig. 1(b). The origin of these peaks can be understood if one examines the condensate polariton distribution function in the pseudospin space. This distribution function is shown in Fig. 2 for four values of the pump. One can see that the in-plane component of pseudospin $S_y$ is strongly suppressed for several values of $S_z$ (see Fig. 2(d) for $P = 3P_{th}$). This happens as a result of spin-anisotropy of polariton-polariton interactions. Indeed, the interaction Hamiltonian (2) can be re-written as $H_{int} = (1/4)\left(\alpha_1 + \alpha_2\right)n^2 + (\alpha_1 - \alpha_2)S_z^2$, and the third term in (1) produces the self-induced Larmor precession [27] of pseudospin around $\hat{z}$-axis. The frequency of this precession is proportional to $S_z$. For a fixed $n$ the in-plane component of pseudospin is averaged to zero for the values of $S_z$ that give full $2\pi$ rotation. In reality the condensate occupation is not fixed and fluctuations also exist, 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 pumping.

**Relaxation of the order parameter.**—The results obtained above are valid for the case of short condensate lifetime $\Gamma_c^{-1}$ compared to the typical relaxation times of the order parameter. In this subsection we show that fast relaxation modifies qualitatively the polarization properties of the polariton condensate. Relaxation not only smears out the Larmor rings discussed above but also favors a linear polarization of the condensate.

Below we consider the simplest form of the relaxation term, namely, model A also referred to as the Landau-Khalatnikov or Onsager model [19]. In this model, the relaxation term in Eq. (4) is written as $\mathcal{R}_s = \delta H_{int}/\delta \psi^*_s$. 

![FIG. 2: Showing the distribution function in normalized pseudospin space, $s = 2S/\pi$, for different values of the excitation pump. Far above the threshold the formation of the Larmor rings is seen.](image-url)
This term favors relaxation of the order parameter to minimize the polariton-polariton repulsion energy. This repulsion energy has its minimum if half of the polaritons in the condensate have their spins oriented up, whilst the other half have their spins down, corresponding to a suppression of the z component of the pseudospin vector and the formation of a linearly polarized condensate. It should be noted that the relaxation term we consider does not conserve the number of polaritons in the condensate. In this way, the condensate depletion and leakage of polaritons from the condensate are implied by the present model.

The distribution functions of polaritons in the normalized pseudospin space for different pump intensities are shown in Fig. 3. One can see that the circular polarization is rapidly lost by the condensate as the pumping intensity increases. The characteristic oscillations of the average linear polarization of the condensate as a function of its circular polarization degree seen in Fig. 2 vanish once the relaxation of the order parameter is switched on. The distribution of polariton condensates in the pseudospin space changes from a sphere to torus and further approaches a flat ring shape as the pumping power increases. The condensate develops a strong spontaneous linear polarization at strong pumping. The build-up of the linear polarization as a result of the polariton BEC has been experimentally observed [1, 2], while in these works the orientation of linear polarization plane has been pinned to one of the crystal axes of the structure due to some intrinsic anisotropy of the samples [1]. Further experimental research is needed in order to evidence the stochastic vector polarization of a polariton condensate.

Conclusions.—We have described theoretically the spontaneous build-up of the vector order parameter in the course of polariton BEC. The build-up of the order parameter manifests itself in formation of the stochastic vector polarization of the condensate. This polarization is correlated with the second order coherence of the condensate. Its statistical distribution depends on the mechanism of polarization relaxation in the polariton system.

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FIG. 3: Showing the effect of relaxation of the order parameter on distribution function in normalized pseudospin space, $s = 2S/\pi$. Contrary to the case without relaxation (Fig. 2) a linear polarization is formed for strong pump above threshold. The relaxation parameter $\gamma = 1 \text{ meV}^{-1} \text{ ps}^{-1}$.

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