Effects of Bose-Einstein condensation of exciton polaritons in microcavities on the polarization of emitted light

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It is shown theoretically that Bose condensation of spin-degenerated exciton polaritons results in spontaneous buildup of the linear polarization in emission spectra of semiconductor microcavities and therefore that linear polarization is a good order parameter for the polariton Bose condensation under unpolarized pumping. If spin degeneracy is lifted, an elliptically polarized light is emitted by the polariton condensate. The main axis of the ellipse rotates in time due to self-induced Larmor precession of the polariton condensate pseudospin. The polarization decay time is governed by the dephasing induced by the polariton-polariton interaction and is strongly dependent on the statistics of the condensed state. If the elliptical polarization preexists in the system as a result of pumping, the lifetime of the linear part of the polarization is also extremely sensitive to the degree of circular polarization induced in the system by pumping. This decay time can be used to measure the coherence degree of the condensate as a function of the polarization of the emitted light, as opposed to more conventional but harder particle counting experiments of the Hanbury Brown-Twiss type.

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I. INTRODUCTION

Exciton polaritons in microcavities[1] are promising candidates to display Bose-Einstein condensation (BEC) in a condensed matter system at high temperature, owing to their unique properties borrowing from their constituents: excitons and photons. From the former they inherit efficient scattering mechanisms, from the latter a very small effective mass and macroscopic coherence length; they further allow convenient probing of the system by simple optical means since cavity polaritons tunnel through the Bragg mirrors and turn into photons whose statistics is identical to that of the polaritons in the condensate. From both, they retain good bosonic behavior at low density.

Existing difficulties in experimental demonstration of polariton BEC are essentially linked to the short lifetime of these particles and the existence of a bottleneck of relaxation when approaching the ground state where condensation is to take place.[2] In this connection an important issue of debate is an unarguable experimental evidence that a condensate has formed in the system. Though the bosonic behavior of polaritons is almost unanimously reckoned after reports of stimulated scattering, narrowing of the photoluminescence line or superlinear intensity of emission,[3–5] for the sake of BEC, these are hints at best which do not allow any quantitative measurement of its coherence. Much progress was realized by Deng *et al.*[6] who measured the zero time delay second-order coherence $g_2(0)$ of the hypothetical condensate. This parameter equals 1 for a coherent state—which is the limiting case for a perfect condensate of noninteracting particles—and 2 for a thermal state, where particles have no phase relationship whatsoever. Though this parameter is a good measure of the coherence degree of a single mode condensate, it is difficult in the case of polaritons to measure experimentally with the standard technique of Hanbury Brown-Twiss (HBT) counting experiments. The Deng *et al.* experiment has not yet been reproduced by other groups and their positive result, reporting a decrease of $g_2(0)$ from 1.8 down to about 1.5 with an increase of pumping, as well as its accuracy, remains to be confirmed. Moreover, strictly speaking Bose condensation is a phase transition linked with the spontaneous symmetry breaking of gauge invariance, that is, with appearance of a well-defined phase in the system, which cannot be evidenced by HBT experiments. Such a phase transition manifests itself in the spontaneous appearance of a nonzero, long living order parameter of the condensate which can be interpreted as an average complex amplitude of the field inside the cavity.

On the other hand, while the quantum properties of the light emitted by a polariton condensate have been addressed theoretically in a number of publications,[7–14] all these works ignored the polarization of cavity modes. Recent experiments have shown that the energy relaxation of polaritons is polarization dependent and that spin dynamics in microcavities is extremely rich and complicated.[11,15–17]

In this paper we propose a simple experimental method to evidence the appearance and survival of the order parameter—that is, of the phase—of a condensate made of interacting polaritons: We show that spontaneous symmetry breaking in an ensemble of polaritons manifests itself in a dramatic change of the linear polarization degree of the light emitted by the cavity and the lifetime of this polarization depends strongly on the nature of the polariton state. Of course, light emitted by microcavities can have a nonzero linear polarization degree without Bose condensation: For instance if the microcavity is excited by a polarized light, the pump-induced polarization can reappear in the ground state. We first consider this situation in opposition to the subsequent case where the polarization builds up along with the condensate, being initially zero and thereby providing an order parameter for Bose condensation. In the presence of preexisting correlations, the linear polarization of emission does not require a definite phase relationship between spin-up and spin-down condensates. For instance, thermal light, with no phase whatsoever, can be polarized. We shall suppose for this case that correlations between the two condensates exist from pumping constraints and thus consider at this stage the effect of statistics (or second-order coherence) only. Namely, we shall consider an elliptically polarized resonant pump in the more general case—which can degenerate to the cases of linear or circular polarization—that injects in the system correlated populations of spin-up and spin-down polaritons. We assume they retain their correlations while relaxing toward the ground state, which is the case if the spin-lattice relaxation is negligible. Therefore we refer to an experimental geometry close to that of Ref. 15 where polarized polaritons were created by resonant pumping at an oblique angle. In the isotropic microcavity the nonresonant circularly polarized pumping (as in Ref. 16) does not create correlations between spin-up and spin-down components of the polariton condensate in the ground state, thus it does not allow for probing of the polariton statistics by linear polarization measurements which is the main subject of this paper. We emphasize that in all cases we consider pulsed excitations, which allow for the measurement of time-resolved emission of the microcavity and study the dynamics of the polarization of the polariton condensate. We demonstrate theoretically that the linear polarization degree of the light emitted by the cavity and especially its lifetime depend sensibly on the statistics of the polariton state, and therefore on $g_2(0)$. Again, it does not depend on the off-diagonal elements of the density matrix, so that a pure coherent state with a well-defined phase on the one hand, and a so-called randomly phased coherent state with same statistics (Poissonian) but no phase on the other hand, will display the same linear polarization.

The phase comes into play when the two fractions of the condensate build up independently, which allows the characterization of the condensation beyond merely particle number statistics. The observation of such an order parameter is difficult if the measurements are performed on a purely circularly polarized state. On the other hand, if two spin-polarized condensates coexist without *a priori* correlations, their interferences give rise to a very particular temporal dependence of linear polarization of the emitted light. Although for the sake of argument we will focus on microcavity polaritons, this applies to any assembly of bosons which combine spin degeneracy and irreversible coupling to the photon field (finite lifetime).

The remainder of the text is organized as follows: in Sec. II we lay down the formalism which relates the pseudospin of ground state polaritons to their quantum state. Our model system is a microcavity pumped out of resonance and incoherently by a polarized or unpolarized pulsed light source. We do not discuss—beyond some short comments postponed at the end of the text—the dynamics of the polariton condensate formation which has already been described elsewhere.[8] Our goal is to describe the time evolution and dephasing of the condensate (and therefore of the linear polarization) versus its coherence degree. The ground state is populated due to both stimulated scattering and spontaneous scattering of polaritons from the upper states. The spontaneous process is responsible for dephasing of the condensate which results in a decay of the order parameter. Its rate is given by $D \approx \Gamma_0/2n_0$[14] where Γ_0 is the radiative broadening and n_0 the population of the condensate. When n_0 is large, this dephasing becomes negligible compared with the energy shifts and the energy broadenings induced by the polariton-polariton interaction, as we show below. We propose a model Hamiltonian for this system and formulate approximations which allow us to integrate it analytically. In Sec. III we discuss in greater detail the notion of coherence degree in a single mode condensate and we introduce a family of states with a varying degree of coherence which are the foundation for the subsequent analysis. In Sec. IV we merge the results from previous considerations to show how one can extract accurate values of $g_2(0)$ from a simple time resolved linear polarization measurement; this is the case where correlations induced by pumping provide the system with a polarization from the start. In Sec. V we study the case where no correlation exists *a priori* in the system. We show how the linear polarization maps to the BEC order parameter and we study its dynamics.

II. FORMALISM

We consider a couple of energy-degenerated spin-up and spin-down quantum states occupied by interacting exciton polaritons, which behave as ideal bosons. Spin-up and spin-down states correspond, respectively, to right- and left-circular polarization of the emitted light. We study the importance of dephasing induced by polariton-polariton interactions in the ground state (weakly depleted condensate) with a general Hamiltonian for interacting spin-polarized particles with two projections of spin.[17] To draw analytical results we neglect the lifetime, the scattering toward spin-forbidden ("dark") exciton states, radiative decay, and spin-lattice relaxation. Excited states with a much longer lifetime keep constant populations on the time scale of the ground state dynamics and contribute a small dephasing by spontaneous emission in the condensate of particles with a random phase. This dephasing will be accounted for through the initial conditions, as its time scale is negligible in comparison to the much quicker dynamics caused by the strong dephasing from interactions in the condensate. With these assumptions the Hamiltonian reads in terms of the annihilation operators $a_{0\downarrow}$ for spin-down and $a_{0\uparrow}$ for spin-up polaritons in the ground state

$$H = \varepsilon(a_{0\uparrow}^\dagger a_{0\uparrow} + a_{0\downarrow}^\dagger a_{0\downarrow}) + W_1(a_{0\uparrow}^\dagger a_{0\uparrow}^\dagger a_{0\uparrow} a_{0\uparrow} + a_{0\downarrow}^\dagger a_{0\downarrow}^\dagger a_{0\downarrow} a_{0\downarrow}) + W_2 a_{0\uparrow}^\dagger a_{0\uparrow} a_{0\downarrow}^\dagger a_{0\downarrow}. \quad (1)$$

The bare polariton energy is ε and pairwise interaction with same (resp. opposite) spin has interaction constant W_1 (resp. W_2). In general, $W_1 \neq W_2$ and often they even have an opposite sign, reflecting the fact that while polaritons with parallel spin repel each other, polaritons with opposite spins may form a bound state, called a *bipolariton*.[18] It has been shown theoretically[19] that $|W_2| \ll |W_1|$ and experimentally[20] that $|W_2| \approx 0.04|W_1|$. In Ref. 19, $|W_1|$ is estimated for the exciton-exciton interaction as

$$|W_1| = 6E_b a_B^2/\mathcal{S} \tag{2}$$

where E_b is the exciton binding energy, a_B the exciton Bohr radius, and $\mathcal{S}$ the surface of the condensate, given to good approximation by the size of the exciting laser spot.

The intensity of right (resp. left) circularly polarized light $\langle a_{0\uparrow}^\dagger a_{0\uparrow}\rangle$ (resp. $\langle a_{0\downarrow}^\dagger a_{0\downarrow}\rangle$) is fixed by initial conditions, unlike $S \equiv a_{0\uparrow} a_{0\downarrow}^\dagger$ which does not commute with Eq. (1) and has a dynamics given by Heisenberg equation

$$i\hbar \dot{S} = [S,H] = V(a_{0\uparrow}^\dagger a_{0\uparrow} - a_{0\downarrow}^\dagger a_{0\downarrow} + 1)S, \tag{3}$$

where $V \equiv 2W_1 - W_2$. Operator S is the ladder operator $S_x + iS_y$ for operators $S_x \equiv \Re a_{0\uparrow} a_{0\downarrow}^\dagger$, $S_y \equiv \Im a_{0\uparrow} a_{0\downarrow}^\dagger$ and $S_z \equiv a_{0\uparrow}^\dagger a_{0\uparrow} - a_{0\downarrow}^\dagger a_{0\downarrow}$, which follow a spin-half algebra. For this reason S is called the *pseudospin*. It is a powerful representation for two-levels systems which allowed many insights into the polaritons spin dynamics.[21] The in-plane components of the pseudospin characterize correlations that exist between spin-up and spin-down condensates. Intensities of linearly polarized components of the emitted light are linked to the pseudospin as follows:

$$I_\leftrightarrow = \frac{n_0}{2} + \langle S_x\rangle, \quad I_\updownarrow = \frac{n_0}{2} - \langle S_x\rangle, \tag{4}$$

where $n_0 \equiv \langle a_{0\uparrow}^\dagger a_{0\uparrow} + a_{0\downarrow}^\dagger a_{0\downarrow}\rangle$ is the total (constant) number of particles, and the degree of linear polarization ρ_l follows as

$$\rho_l = \frac{2|\langle S\rangle|}{n_0}, \tag{5}$$

which makes clear that spin interactions between the many particles of a condensate in the ground state hypothetically yield some dynamics worth studying of the linearly polarized components, which we now endeavor to prove. Since $a_{0\downarrow}^\dagger a_{0\downarrow}$ and $a_{0\uparrow}^\dagger a_{0\uparrow}$ are two constants of motion under Eq. (1), in Heisenberg picture Eq. (3) is integrated straightforwardly

$$S(t) = e^{-iVt/\hbar} \exp\left(\frac{iVt}{\hbar}(a_{0\downarrow}^\dagger a_{0\downarrow} - a_{0\uparrow}^\dagger a_{0\uparrow})\right) S(0). \tag{6}$$

$S(t)$ is the projection of pseudospin on its initial direction. The factor $V(\langle a_{0\downarrow}^\dagger a_{0\downarrow}\rangle - \langle a_{0\uparrow}^\dagger a_{0\uparrow}\rangle)$ is the energy splitting between right- and left-circularly polarized condensates which arises if their populations are not equal. This splitting, also referred to as optically induced Zeeman splitting, has been theoretically analyzed[22] and experimentally observed.[23] The pseudospin operator thus rotates at a speed given by the energy splitting between the two condensates. The remarkable feature of this result arises when we move to quantum averages over possible states of the condensate. Before we return to this point in Secs. IV and V, we first explain a gamut of states which characterize the ground state as its coherence degree varies from zero (thermal state) to one (coherent state).

III. SECOND-ORDER COHERENCE

The most relevant quantity to describe with a single scalar quantity the quantum state of a single mode is the so-called second-order correlator g_2 defined as

$$g_2(t,\tau) \equiv \frac{\langle a_0^\dagger(t) a_0^\dagger(t+\tau) a_0(t+\tau) a_0(t)\rangle}{|\langle a_0^\dagger(t) a_0(t)\rangle|^2} \tag{7}$$

for spatially homogeneous cases. We consider here the case of $g_2(t,0)$ only (zero-delay second order correlation function) since this is for zero delay that the field statistics are most clearly imprinted in this quantity: For infinite delays all particles become uncorrelated and g_2 is 1, regardless of the underlying quantum state. At zero delay, however, g_2 equals 1 for the case of a coherent state, while it grows to 2 for so-called thermal states, exhibiting the bunching effect typical of incoherent light.[24] g_2 is generally measured by HBT experiments which are quantum optical in nature: They require single photon detections at the same time, from which one infers statistical correlations. This is a rather delicate experimental measure, but from the mathematical point of view, $g_2(0)$ is merely computed from diagonal elements $p(n)$ of the density matrix

$$g_2(0) = \frac{\sum_{n=0}^{\infty} n(n-1)p(n)}{[\sum_{n=0}^{\infty} np(n)]^2}. \tag{8}$$

From the above expression, it is straightforward to express $g_2(0)$ as a function of the first two moments of $p(n)$, the mean $n_0 = \langle a_0^\dagger a_0\rangle$ and the variance $\mathrm{Var}(a_0^\dagger a_0) = \sigma^2$

$$g_2(0) = 1 + \frac{\sigma^2 - n_0}{n_0^2}. \tag{9}$$

In Sec. IV we shall see that to a very good approximation the quantum average of S is also a function of the first two moments of $p(n)$ and thus a function of g_2. In the meantime we discuss the various statistics of interest in our case. We have already spoken of the two extremes, namely, the coherent case and the thermal case. The coherent case has Poisson statistics

$$p_{\mathrm{coh}}(n) = e^{-n_0}\frac{n_0^n}{n!}, \tag{10}$$

here of course $\sigma^2 = n_0$. The distribution is sharply peaked about its mean, with small fluctuations in particle number corresponding to the smallest quantum uncertainty allowed for a state without amplitude squeezing. It is at the same time the most classical state of the quantum realm (mapping as closely as allowed by quantum mechanics to a monochromatic wave) and the ideal BEC picture for noninteracting particles. This would be the state emitted by an ideal, noiseless laser far above its threshold. Its second-order coherence correlator $g_2(0)$ is 1. On the opposite, the thermal state, with

$g_2(0)=2$, has exponentially decreasing statistics

$$p_{\mathrm{th}}(n) = \frac{n_0^n}{(1+n_0)^{n+1}}. \tag{11}$$

Now $\sigma^2=n_0^2+n_0$ and particles fluctuate wildly in the system with occupancy of highest probability for the vacuum. For temperatures at which microcavities are operated, this precludes high occupancy numbers, whereas we investigate condensates which imply such high populations in a single state. We will briefly consider this case as a limiting case of mathematical interest but bear in mind that it is not realistic.

The physically relevant case is that of an essentially coherent state, say with n_c particles, which is dephased by a superimposed fraction of a thermal state, with n_t particles. Note that the denomination of "thermal" state does not imply thermalization *per se*, but rather dephasing of the kind which is best and most commonly illustrated by a field in thermal equilibrium, that is with random phase and amplitude which stems from a random walk (Gaussian). The small fraction of this so-called thermal state which broadens the coherent state is caused by various dephasing mechanisms, like spontaneous diffusion from excited states. Such states are well known to describe laser light above threshold.[25] For excitons or polaritons, they have been obtained solving dynamically quantum Boltzmann master equations.[8,26] We will conveniently refer to such particles as coherent and incoherent, respectively, though of course once the coherent fraction and the thermal fraction are merged, a particle does not belong any longer to a part of this decomposition but is indistinguishable from any other of the lot. This is just a vivid picture to describe a collective state which has some phase and amplitude spreading. We define the *second-order coherence degree* χ as the ratio of the number of coherent particles over the total number of particles

$$\chi = \frac{n_c}{n_c+n_t} \tag{12}$$

with $n_c+n_t=n_0$. The density matrix of such a state is easily built from Glauber's P representation[27] of the density matrix, i.e., the "weighting factor" of ρ in the basis of coherent states $|\alpha\rangle$

$$\rho(t) = \int_{\mathbb{C}} P(\alpha,\alpha^*,t)|\alpha\rangle\langle\alpha| d\alpha d\alpha^*. \tag{13}$$

The P function for the superposition of two uncorrelated fields is given by the convolution of their P functions, which are a δ function for the coherent state and a centered Gaussian for a thermal state.[24,28] As a result, a whole gamut of states with some degree χ of coherence is modeled after off-centered Gaussians, where the mean yields the coherent fraction n_c/n_0 and where the spread yields the thermal fraction n_t/n_0

$$P_{\mathrm{coth}}(\alpha,\alpha^*) = \frac{1}{\pi n_t} e^{-|\alpha-\sqrt{n_c}e^{i\varphi}|^2/n_t}. \tag{14}$$

We have subscripted with "coth" these states which are a mixture of *co*herent and of *th*ermal states. Here φ is the mean phase of the condensate. The order parameter of the condensate is $\sqrt{n_c}e^{i\varphi}$ and is zero if $n_c=0$, which is the thermal case. The mean of P_{coth} is of course n_0. Its variance needs to be computed

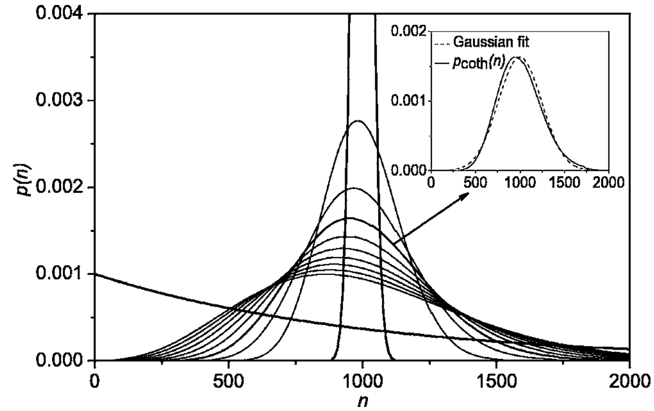

FIG. 1. Probability distributions $p_{\mathrm{coth}}(n)$ of condensates with $n_0=10^3$ and with various degree of coherence, namely, with χ running from 99% (sharper thin curve) to 91% (flatter thin curve) together with limiting cases of coherent (100%) and thermal (0%) states, in thick lines. The Poisson distribution of the coherent state assumes maximum value $p_{\mathrm{coh}}(10^3)\approx 0.013$, more than three times higher than is visible. In inset is shown for $\chi=97\%$ the Gaussian fit used to compute Eq. (31). The approximation is better for higher χ.

$$\mathrm{Var}(P_{\mathrm{coth}}) = n_0 + n_t^2 + 2n_c n_t, \tag{15}$$

and allows us to link χ and $g_2(0)$ as

$$g_2(0) = 2-\chi^2. \tag{16}$$

From Eqs. (13) and (14) one can extract the statistics needed to compute $\langle S(t)\rangle$

$$p_{\mathrm{coth}}(n) = \int_{\mathbb{C}} P_{\mathrm{coth}}(\alpha,\alpha^*)|\langle n|\alpha\rangle|^2 d\alpha d\alpha^*, \tag{17}$$

which evaluates to

$$p_{\mathrm{coth}}(n) = \exp\left(-\frac{n_0\chi}{1+n_0(1-\chi)}\right)\frac{[1+n_0(1-\chi)]^{n+1}}{[n_0(1-\chi)]^n} \times \mathrm{L}_n\left(-\frac{\chi}{(1-\chi)[1+n_0(1-\chi)]}\right) \tag{18}$$

where L_n is the nth Laguerre polynomial. This distribution is plotted in Fig. 1 for values of χ ranging from 0.91 to 0.99 by step of 1%, also with the two limiting cases of the pure coherent state ($\chi=1$) and the thermal state ($\chi=0$). As one can see, this distribution very quickly broadens for small deviations from the coherent state, and becomes thermal-like, with huge fluctuations of particle numbers, even for a neatly dominant proportion of coherent particles. In the next sections we show how this results in sharp dependency of polarization on coherence.

IV. POLARIZATION DYNAMICS FOR CORRELATED CONDENSATES

We now show how $\langle S(t)\rangle$ depends crucially on the quantum state, specified in its more general form by a density matrix ρ of the spin-degenerated condensate, which is time independent in the Heisenberg picture and thus fully specified by its initial condition. We first investigate the case where the two condensates are correlated from pumping condition, in the next section we turn to the case where the pumping is unpolarized and where polarization buildup can be used as an evidence of Bose condensation. We therefore address the experimental situation of a polariton condensate excited by fully polarized nonresonant pumping. In case of no spin relaxation, the reservoir of polarized polaritons formed at the excitonic part of the lower polariton dispersion branch feeds the condensate formed at the ground state, so that the condensate lifetime exceeds by a few orders of magnitude the single polariton lifetime and can be assumed to be infinite on the time scale of our interest.

As we consider elliptically polarized pumping in the general case, it is advantageous to work in the basis of elliptically polarized states. A polariton with circular polarization degree given by $P\equiv\cos^2\theta-\sin^2\theta$ is the coherent superposition of a spin-up polariton with probability $\cos^2\theta$ and of a spin-down polariton with probability $\sin^2\theta$, therefore, its quantum state can be created from the vacuum $|0,0\rangle$ (zero spin-up and zero spin-down polaritons) by application of the following operator:

$$|1,\theta,\phi\rangle \equiv (\cos\theta a_{0\uparrow}^{\dagger} + e^{i\phi}\sin\theta a_{0\downarrow}^{\dagger})|0,0\rangle. \tag{19}$$

Here we also took into account the angle ϕ of in-plane orientation of the axis of the polarization ellipse, which however plays no role in what follows. This defines $a_{\theta,\phi}^{\dagger}$ the creation operator for an elliptically polarized polariton as

$$a_{\theta,\phi}^{\dagger} \equiv \cos\theta a_{0\uparrow}^{\dagger} + e^{i\phi}\sin\theta a_{0\downarrow}^{\dagger}. \tag{20}$$

The superposition of n such correlated polaritons is obtained by recursive application of the creation operator

$$|n,\theta,\phi\rangle = a_{\theta,\phi}^{\dagger n}|0\rangle = \frac{1}{\sqrt{n!}}(\cos\theta a_{0\uparrow}^{\dagger} + e^{i\phi}\sin\theta a_{0\downarrow}^{\dagger})^n|0,0\rangle, \tag{21}$$

which we have normalized (here $|0\rangle$ is the vacuum in the space of elliptically polarized states). Writing the density matrix in this basis, one obtains

$$\langle S(t)\rangle = \sum_{n,n'} \rho_{n,n'}\langle n,\theta,\phi|e^{-iVt/\hbar} \times\exp\left[\frac{iVt}{\hbar}(a_{0\downarrow}^{\dagger}a_{0\downarrow} - a_{0\uparrow}^{\dagger}a_{0\uparrow})\right]S(0)|n',\theta,\phi\rangle. \tag{22}$$

Simple but lengthy algebra yields for the matrix element (details of the derivation are given in the Appendix)

$$\langle n,\theta,\phi|\exp\left[\frac{iVt}{\hbar}(a_{0\downarrow}^{\dagger}a_{0\downarrow} - a_{0\uparrow}^{\dagger}a_{0\uparrow})\right]S(0)|n',\theta,\phi\rangle = s_0 n\Theta(t)^{n-1}\delta_{n,n'}, \tag{23}$$

with $s_0 = 1/2\sin 2\theta e^{-i\phi}$ is the in-plane pseudospin of a single elliptically polarized polariton, and where we introduced as a shortcut

$$\Theta(t) \equiv \cos^2\theta e^{-iVt/\hbar} + \sin^2\theta e^{iVt/\hbar}, \tag{24}$$

from which follows the direct connection between the pseudospin, or linear polarization, and the statistics $p(n)\equiv\rho_{n,n}$ of the condensate

$$\langle S(t)\rangle = s_0\langle n\Theta^{n-1}\rangle, \tag{25}$$

where the right-hand side average is over $p(n)$. Note that $\langle S(t)\rangle$ depends only on diagonal elements of the density matrix, that is, it depends on the statistics only and does not reflect the behavior of the phase (apart from s_0 which is time independent). Let us now evaluate the in-plane pseudospin with the statistics introduced in the previous section. The results that are shown correspond to a typical CdTe microcavity with a lateral size of $L=60$ μm, an exciton binding energy $E_b=25$ meV, an exciton Bohr radius $a_B=40$ Å and with the average number of polaritons in the condensate $n_0\approx 10^5$. This gives the interaction strength $V\approx 10$ neV [obtained by multiplying formula (2) by the exciton fraction of the polariton ground state, which is $1/2$ at zero detuning].

In the pure coherent case (10), the average pseudospin (25) is easily computed as

$$\langle S(t)\rangle = \langle S(0)\rangle\exp\left[n_0\left(\cos\frac{Vt}{\hbar}-1\right)\right]\exp\left[in_0P\sin\frac{Vt}{\hbar}\right] \tag{26}$$

with $\langle S(0)\rangle\equiv s_0 n_0$ the initial in-plane pseudospin and $P\equiv\cos^2\theta-\sin^2\theta$ the circular polarization degree. The in-plane polarization oscillates with a period

$$T_{\mathrm{coh}} = \frac{2\pi\hbar}{n_0VP} \tag{27}$$

given by the energy splitting between the circularly polarized eigenstates n_0VP. If the spin degeneracy is not lifted, $\theta=\pi/4$ and the polarization axis does not rotate, only dephasing takes place. This is the case of a purely linearly polarized condensate made up of two completely correlated spin-up and spin-down condensates of equal populations. If the condensate is elliptically polarized (with different average numbers of particles in its spin-up and spin-down components) the main axis of the ellipse rotates with time: This is self-induced Larmor precession. The projection of the pseudospin on its initial direction oscillates in this case. The amplitude of these oscillations decays like $e^{-t^2/\tau_{\mathrm{coh}}^2}$ with time constant

$$\tau_{\mathrm{coh}} = \frac{\sqrt{2}\hbar}{\sqrt{n_0}V}. \tag{28}$$

This decay should be followed by a revival after a time $\sqrt{2}\hbar/V$ which for parameters we consider falls in the microsecond range. Thus it cannot be observed since the polarization in the system will be lost because of the weak interactions with polaritons from excited states (which were neglected in this model). In the thermodynamic limit—where $\sqrt{n_0}V$ goes to 0 with the area of the sample occupied by the condensate going to infinity, while the polariton density remains constant—the dephasing of the coherent state vanishes, recovering a thermodynamical virtue of BEC in infinite size systems.

In the thermal case (11), the average pseudospin (25) is computed as

$$\langle S(t)\rangle = \frac{\langle S(0)\rangle}{\{n_0[1-\Theta(t)]+1\}^2}. \tag{29}$$

The polarization decay time is given in this case by

$$\tau_{\rm th} = \frac{\sqrt{2}\hbar}{n_0 V}, \tag{30}$$

which is about 30 ps for the parameters of our model. As the decay time is shorter than the period of the oscillations, none can be observed in this case. Contrary to the coherent case, the broadening does not vanish in the thermodynamic limit. This shows that a linear polarization in the thermal state is impossible. In reality, a pure thermal state is never realized but if the thermal fraction is nonzero then the broadening does not vanish either in the thermodynamic limit.

Now we turn to the general case (18). In principle one can compute numerically $\langle S(t)\rangle$ and in this way extract the period of polarization oscillation and decay time. We repeat however that the region of interest is close to a coherent state where a minute variation of χ results in important changes of the statistics. Observe also that $p_{\rm coth}$ in this region can be approximated by a Gaussian. The inset in Fig. 1 shows the quality of this approximation for $\chi=97\%$ which we will see is already far enough from the coherent states for interesting effects to have already been observed. We therefore replace the awkward exact distribution (18) by a Gaussian which mean and variance are given by the first two moments of Eq. (18), that is, n_0 and $\sigma^2=n_0+n_0^2(1-\chi^2)$. This allows evaluating Eq. (25) in the continuous limit $s_0\langle x\Theta^{x-1}\rangle$ to obtain an analytical expression in a neighborhood of the coherent state

$$\langle S(t)\rangle = \langle S_0\rangle \exp\left(n_0 \log\Theta(t) + \frac{1}{2}\sigma^2[\log\Theta(t)]^2\right) \tag{31}$$

after neglecting logarithmically small values. Confronting this expression with numerical computations proves it to be sound even far away from the coherent state. In the limit $t \ll \hbar V$, Eq. (31) reads to order two in time

$$\langle S(t)\rangle \approx \langle S_0\rangle \exp(-t^2/\tau^2)\exp(i2\pi t/T_{\rm coh}) \tag{32}$$

with a decay time τ given by

$$\tau = \frac{\sqrt{2}\hbar}{V\sqrt{n_0+n_0^2P^2(1-\chi^2)}}, \tag{33}$$

while the period of polarization oscillation keeps the same value (27) independent of the coherence degree [close to the pure coherent state, this comes from approximating to n_0 the value at which $p_{\rm coth}(n)$ is maximum]. Most striking effects therefore belong with the lifetime (33) which is the central result of this section. Figure 2 shows the decay time of the polarization versus the coherence degree of the condensate for different linear polarization degree, for the structure already described. Note the peculiar influence of the circular polarization degree P. The decay time τ depends strongly on it once coherence starts to decrease from one but is otherwise unaffected in the pure coherent case. This polarization dependence is not recovered in the limiting thermal case [cf. Eq. (30)], where Eq. (33) does not apply anyway and which is not physical. Apart from P, however, the formula is qualitatively right. Pinning the polarization from pumping one can thus accurately measure $g^2(0)$. This measure requires no quantum optical setup and can be fully realized thanks to time-resolved polarized photoluminescence. The accuracy is very good thanks to steep variation for well-chosen polarization. The population n_0 can be determined from the purely linearly polarized case, where the coherence degree plays no role. Excited states may also strongly contribute to the dephasing of the condensate but our present theory does not describe this effect.

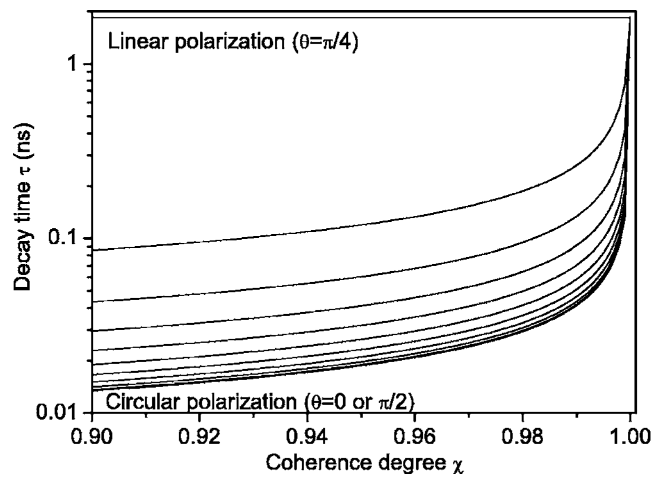

FIG. 2. Decay time τ of the polarization in the case of polarized pumping as given by Eq. (33), as a function of the coherence degree χ in a neighborhood of the coherent state, for $P=\cos^2\theta-\sin^2\theta$ ranging from 0 (upper envelope) to ± 1 (lower envelope) for θ varying by steps of 1/40 rad. For a pure coherent state, the lifetime does not depend on the polarization. By adjusting this polarization from pumping, one can tune the steepness of decay and therefore the accuracy of the measurement. For the linear polarization, τ depends on θ on the fourth order of time only, and the change of decay with χ is therefore not experimentally observable in this case. This can be used to measure population or interaction strength, independently of the quantum state of the condensate.

V. LINEAR POLARIZATION BUILDUP AS A SIGNATURE OF SYMMETRY BREAKING

The previous analysis pertained to the case where correlations existed between the two fractions of the condensate, as is the case when polaritons are created by a polarized pumping. We now turn to the case where there are no correlations, that is, to the case of unpolarized nonresonant pumping. This brings the important feature that the appearance of the order parameter in the condensate leads to the spontaneous buildup of linear polarization. This polarization appears in the system without preexisting there. If the populations of each spin projection are equal, its in-plane orientation is constant in time, but randomly changes from experiment to experiment in isotropic system. This linear polarization bears much with a symmetry breaking and as we shall see is in fact mapped to the product of the order parameters $\langle a_{0\downarrow}\rangle$ and $\langle a_{0\uparrow}\rangle$. In this sense, it serves as an order parameter for Bose condensation of polaritons, that is, of particles with a spin degree of freedom (so that the interference can take place) and finite radiative lifetime (so that the effect on polarization of the light emitted can be observed). In this respect, this simple effect has no possible realization with atoms.

In general the spin degeneracy is significantly lifted by fluctuations feeding spin-up and spin-down condensates with unequal populations. For classical particles these fluctuations would yield a mean imbalance of $\sqrt{n_{0\uparrow}+n_{0\downarrow}}$ particles between the two condensates with $n_{0\uparrow}$ spin-up and $n_{0\downarrow}$ spin-down particles and consequently the circular polarization degree $\rho_c=(n_{0\uparrow}-n_{0\downarrow})/(n_{0\uparrow}+n_{0\downarrow})$ would vanish like the inverse square root of the occupation number. However, because of stimulation, the probability to reach one condensate or the other depends on respective populations in such a way as to strengthen the more populated state, leading to possibly highly degenerated configurations. The probability for a particle to join the condensate with $n_{0\uparrow\downarrow}$ particles is

$$p_{\uparrow\downarrow}=\frac{n_{0\uparrow\downarrow}+1}{n_{0\uparrow}+n_{0\downarrow}+1}. \tag{34}$$

This yields $\langle|\rho_c|\rangle=(2+n_0)/(2+2n_0)$ which is approximately 1/2 for large values of $n_0\equiv n_{0\uparrow}+n_{0\downarrow}$. This corresponds to an elliptically polarized light. Most frequently time-resolved polarization is measured under pulsed excitation and time averaging of the emitted signal over a large number of pulses. In such an experimental configuration, the average linear polarization degree of the emitted light is zero, since it assumes a random value after each excitation pulse. The absolute value of linear polarization should therefore be recorded after each pulse, in order to demonstrate experimentally the Bose condensation of the particles.

The analysis follows the same lines as previously starting with the same Hamiltonian (1) until the time dependent expression (6) for the pseudospin. At this point, evaluation of the average $\langle S(t)\rangle$ differs because the two fractions of the condensate—namely, spin up and spin down—have been formed independently and are not correlated, i.e., the density matrix of the ground state factorizes as $\rho=\rho_\uparrow\otimes\rho_\downarrow$ and consequently

$$\langle S(t)\rangle=e^{-iVt}\mathrm{Tr}[\exp[-iVta^\dagger_{0\uparrow}a_{0\uparrow}]a_{0\uparrow}\rho_\uparrow] \times\mathrm{Tr}[\exp[iVta^\dagger_{0\downarrow}a_{0\downarrow}]a^\dagger_{0\downarrow}\rho_\downarrow]. \tag{35}$$

The initial in-plane pseudospin reads

$$\langle S(0)\rangle=\alpha_\uparrow\alpha^*_\downarrow \tag{36}$$

with the definition of order parameter for each circularly polarized condendate given, as usual, by $\alpha_{\uparrow\downarrow}\equiv\langle a_{0\uparrow\downarrow}\rangle=\mathrm{Tr}(a_{0\uparrow\downarrow}\rho_{\uparrow\downarrow})$. From Eqs. (5) and (36) one establishes an explicit connection between the linear polarization ρ_l and order parameter of BEC defined in the usual way as the system average over the Bose annihilation operator. Indeed appearance of the linear polarization in the condensate is observed only if an order parameter builds up for each of the circularly polarized components. The measurement of the circularly polarized emission gives access to $n_{0\uparrow\downarrow}$ which combined with the measurement of the linear polarization degree gives a measurement of the order parameter. Note that the superposition of two states with a Poisson distribution but no well-defined phase (randomly phased coherent states) does not lead to an in-plane polarization, so that the effect is really associated with the phase, not merely with coherence in the sense of Poisson statistics.

In what follows we compute the time dependence of the in-plane pseudospin versus the coherence degree of the individual condensate using Glauber representation of the density matrix (13) upgraded to describe the spin degree of freedom,

$$\rho_{\uparrow\downarrow}=\int|\alpha_{\uparrow\downarrow}\rangle\langle\alpha_{\uparrow\downarrow}|P(\alpha_{\uparrow\downarrow},\alpha^*_{\uparrow\downarrow})d\alpha_{\uparrow\downarrow}d\alpha^*_{\uparrow\downarrow} \tag{37}$$

with α here characterizing the coherent state $|\alpha\rangle$ (with a given amplitude and phase). From this definition one obtains

$$\langle S(t)\rangle=e^{-iVt}\int P_\uparrow(\alpha_\uparrow,\alpha^*_\uparrow) \times\langle\alpha_\uparrow|e^{-iVta^\dagger_{0\uparrow}a_{0\uparrow}}a_{0\uparrow}|\alpha_\uparrow\rangle d\alpha_\uparrow\int P_\downarrow(\alpha_\downarrow,\alpha^*_\downarrow) \times\langle\alpha_\downarrow|e^{iVta^\dagger_{0\downarrow}a_{0\downarrow}}a^\dagger_{0\downarrow}|\alpha_\downarrow\rangle d\alpha_\downarrow. \tag{38}$$

The initial coherence degree in each of the individual condensates is given by, cf. Eq. (12)

$$\chi_{\uparrow\downarrow}=\frac{|\alpha_{\uparrow\downarrow}|^2}{n_{0\uparrow\downarrow}}=\frac{n_{0\uparrow\downarrow,c}}{n_{0\uparrow\downarrow,c}+n_{0\uparrow\downarrow,t}}=\sqrt{2-g^2_{\uparrow\downarrow}(0)}, \tag{39}$$

with $g^2_{\uparrow\downarrow}(0)$ the second-order coherence of the individual condensates, $n_{0\uparrow\downarrow,t}$ the average numbers of spin-up and spin-down polaritons in the thermal fraction, and $n_{0\uparrow\downarrow,c}=|\alpha_{\uparrow\downarrow}|^2$ the average numbers of spin-up and spin-down polaritons in the coherent fraction, in the sense outlined in Sec. III. Here also $\chi_{\uparrow\downarrow}$ varies between 0 (thermal state) and 1 (coherent state).

Using Eq. (14) for each fraction of the condensate in the above formula, one finds

$$\langle S(t)\rangle=\frac{\langle S(0)\rangle\exp\left(-\frac{n_{0\uparrow,c}\theta}{n_{0\uparrow,t}\theta+1}-\frac{n_{0\downarrow,c}\theta^*}{n_{0\downarrow,t}\theta+1}\right)}{(n_{0\uparrow,t}\theta+1)^2(n_{0\downarrow,t}\theta^*+1)^2} \tag{40}$$

with $\theta \equiv 1-\exp[-iVt]$.

We now consider the likely configuration where the coherence degrees of spin-up and spin-down condensates are equal and given by χ. In the limit $Vt \ll 1$, expression (40) is approximately given by

$$\langle S(t)\rangle = \langle S(0)\rangle \frac{e^{-in_0\rho_c\chi Vt}e^{-\frac{1}{2}(n_0+(1-\chi)(1+\rho_c^2)n_0^2)\chi V^2t^2}}{\left(\frac{1}{2}(1-\chi)(1+\rho_c)n_0 iVt+1\right)^2\left(\frac{1}{2}(1-\chi)(1-\rho_c)n_0 iVt-1\right)^2}. \tag{41}$$

The behavior of the pseudospin is dominated by the numerator of Eq. (41) in the vicinity of the coherent case ($\chi \approx 1$) and by the denominator in the opposite limit, close to the thermal case ($\chi \approx 0$). In a narrow region close to full coherence, the pseudospin oscillates in time with a period

$$T_0 = \frac{2\pi}{Vn_0\chi|\rho_c|}. \tag{42}$$

Conversely to the period of oscillations, the amplitude is very sensitive to the coherence degree. The pseudospin decays like $\exp(-t^2/\tau^2)$ with characteristic time

$$\tau = \frac{\sqrt{2}}{V\sqrt{(n_0+(1-\chi)(1+\rho_c^2)n_0^2)\chi}}. \tag{43}$$

This decay is caused by the energy broadening of the state which is induced by the huge thermal fluctuations in particle number which result in fluctuations of energy and hence on destructively interfering oscillations of the Larmor precessions. For the completely coherent case where the fluctuations in the particle number are as small as allowed without squeezing, the decay time is as high as

$$\tau_{\text{coh}} = \frac{\sqrt{2}}{V\sqrt{n_0}}. \tag{44}$$

Note that it increases with L^2 so that the polariton density remains constant, thus the dephasing of a coherent state vanishes in the thermodynamic limit, which again fits well with the classical picture of BEC. However, the presence of even a tiny thermal fraction dramatically reduces the decay time. If $n_0(1-\chi) \gg 1$, the decay times evaluates to

$$\tau = \frac{\sqrt{2}}{Vn_0(1-\chi)}, \tag{45}$$

which is vanishing. Contrary to τ_{coh}, it remains finite in the thermodynamic limit, thus looses the linear polarization of the condensate no matter how small the thermal fraction. Thus, only in a narrow region close to full coherence does the pseudospin exhibit oscillations in time. For values of χ below 85%, there are no observable oscillation and the decay is very fast as well as almost independent of the coherence degree. In the limit of small χ, the pseudospin decays like a Lorentzian

$$|\langle S(t)\rangle| \approx \frac{1}{|1+2i(1-\chi)n_0V|\rho_c|t|}. \tag{46}$$

On the opposite, close to the coherent limit, the pseudospin oscillates with a period given by the energy splitting between the circularly polarized eigenstates in the coherent fraction.

It also depends on the coherence degree but since the range of validity of this formula is for a small domain of χ about one, the period of oscillations is essentially independent of the coherent properties of the state. This period is however sensitive to the circular polarization degree of the condensate which absolute value runs from 0 to 1. The lower limit is reached when the spin degeneracy is not lifted in which case the polarization axis does not rotate and only dephasing takes place. The orientation of linear polarization is random in a system having a perfect in-plane isotropy.

Figure 3 displays the decay time τ as a function of the coherence degree χ. Parameters are still for a CdTe cavity, now with $n_0=10^4$ polaritons in the ground state and $\rho_c=1/2$. The solid-dotted line results from numerical calculations with Eq. (41), estimating the typical lifetime as the time it takes for $|S(t)|$ to decrease by a factor e. This is natural in the limit of coherent states where the decay is exponential. The solid line superimposed is obtained analytically from Eq. (43) which holds over half the defining domain of χ. The curve is displayed dotted below 50% where it looses physical meaning. Past this point, the decay looses its exponential

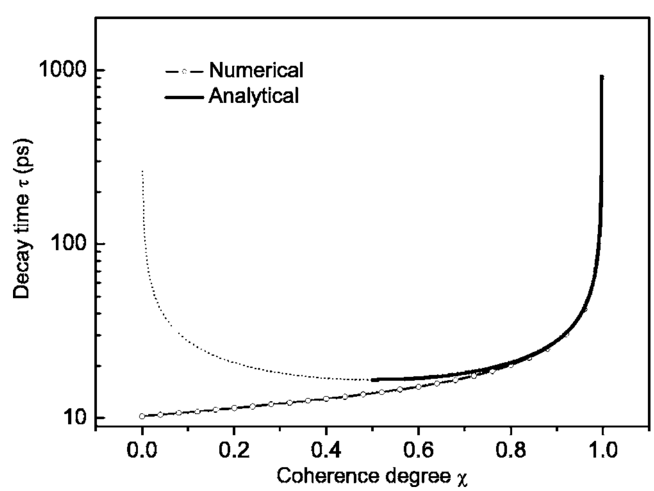

FIG. 3. Decay time of the polarization in the case of unpolarized pumping, with bullets on numerical points and its analytical approximation (43) which holds in the vicinity of coherent cases (solid), here for $\rho_c=1/2$. The unphysical behavior of Eq. (43) out of its range of validity is shown dotted.

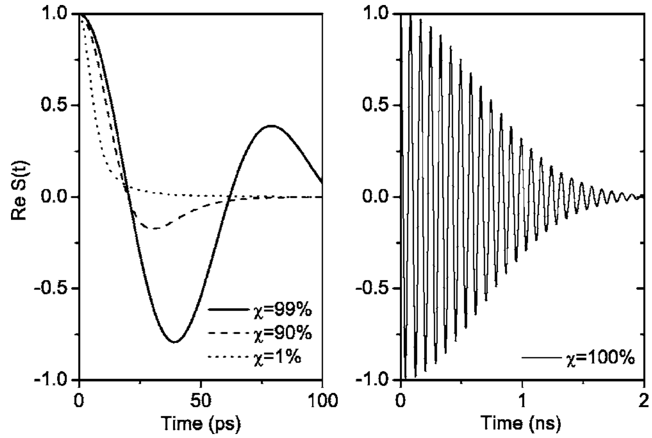

FIG. 4. Decay of the linear polarization in the case of unpolarized pumping for the full coherent case ($\chi=1$) on the right, featuring sustained oscillations, and the impact of thermal contamination ($\chi=0.01$, 0.9 and 0.99) on the left. Note the different time scale (the period is the same on both graphics.)

character to behave according to the denominator of Eq. (46), i.e., approximately like a Lorentzian. Figure 4 shows the decay of the pseudospin in these two opposite regimes. The right-hand side displays the pure coherent case, where many oscillations are sustained for as long as several nanoseconds even though there is a very large number of particles. The decay time caused by spontaneous emission is even longer (few hundreds of nanoseconds in the present case). It is interesting to compare the dephasing time and the typical coherence buildup time, the characteristic time needed for a coherent state to appear after the nonresonant pumping is switched on, which we have found to be, for CdTe microcavities, of the order of a few hundreds of picosecond. This comparison shows that the dephasing induced by the polariton-polariton interaction does not prevent formation of coherent states and therefore symmetry breaking in polariton systems. The left-hand side displays two almost coherent cases where the decay time has drastically decreased because of the phase mismatches brought by the thermal fraction. Also displayed is the thermal case, here for $\chi=1\%$, though this overdamped, nonoscillating decay is characteristic for all cases with $\chi<85\%$. Since the decay time of $|S(t)|$ is very sensitive to the coherent degree of the condensate, it allows for easy and accurate measurements. We also point out that the dephasing time of a single component condensate can be straightforwardly extracted from the present formalism. We do not address specifically this aspect because this quantity is much harder to measure for a single component condensate than for a superposition of two different ones.

As an aside we now comment briefly on connections between the results presented here relating to the dephasing of the condensate and results relating to its buildup, especially those published by the present authors in Ref. 8. The main issue is whether the dephasing time is long enough so that the condensate has time to form before its polarization is irremediably lost. This is an important question for otherwise the predicted effect cannot be observed. Yet its quantitative answer is out of the scope of the present paper, where we have confined the buildup stage to initial conditions. As the dephasing depends on the number of particles in the condensate, one needs to couple the present model to the formalism laid down in Ref. 8, which is not straightforward technically as it doubles the number of equations in an already heavy system. We now offer some qualitative support for the possibility to operate the cavity in a regime where polarization outlives the buildup time. First, one should not directly compare the time scales of the coherence buildup and the polarization dephasing, since the rapid dephasing comes from high occupancy and therefore applies to the fully formed condensate. It is less strong when the latter is in its buildup stage with fewer particles. Second, Ref. 8 also shows that while time scales for coherence and population buildup are the same, coherence nevertheless has a steeper increase than population so that the high coherence and small occupancy favor a qualitative support for an initial state which has a high degree of polarization and of coherence, as we have assumed. That the coherence degree can be as high as we have demanded is a result of our own findings but also of Ref. 26 where—fitting the statistics here obtained with p_{coth}, Eq. (18)—coherence degrees significantly higher than 99% are found. So under the suitable experimental conditions, as coherence buildup and polarization dephasing are largely unrelated and can be tuned independently, one can maximize their respective time scales to offer the best visibility of our effect.

Finally, let us discuss the relevance of the present model to the important case of a polariton laser working in the cw regime. To describe correctly this situation, one should introduce the mechanism of spontaneous symmetry breaking responsible for the buildup of linear polarization in the system. Our present consideration remains valid for estimation of the lifetime of this polarization which is nothing but the coherence time of the polariton laser (directly linked to its coherence length). In cw regime, emission of the polariton laser must be always polarized if the symmetry breaking took place. However, orientation of this spontaneous polarization randomly changes on a time scale given by the decay time we calculated here.

VI. CONCLUSIONS

We have shown how the linear components of polarization of the light emitted by a microcavity-polaritons condensate is intimately related to its coherence and phase property. We showed that the decay time of the linear polarization depends strongly on the circular polarization degree which can be tuned experimentally by the polarization of the pump. In the vicinity of a coherent state with $g_2(0)\approx 1$ the lifetime of the linear polarization becomes orders of magnitude longer than in thermal or mixed states where $g_2(0)>1$. This measure therefore allows an accurate, though indirect, determination of the zero delay second-order coherence. In the case where the pumping light is unpolarized, the spontaneous appearance of linear polarization is a criterion for Bose condensation as it implies a well-defined phased (symmetry breaking) as well as a high degree of coherence (Poisson statistics). The degree of linear polarization depends on the order parameters of the two fractions (spin up and spin down) of the condensate. Its decay time increases with an increase of the degree of coherence of the condensates. In case of a fully coherent state, it is proportional to the square root of the number of polaritons in the ground state. If the polariton condensate is elliptically polarized and in a coherent state, the in-plane component of its pseudospin rotates with a period proportional to the circular polarization degree of the condensate and to the number of polaritons in the ground state. This results in rotation of the main axis of the polarization ellipse of the emitted light. Thus, by measuring time-resolved linearly polarized photoluminescence one can obtain detailed information on population of polariton condensates, their order parameters and coherence degrees. We focused on the ground state only, thereby neglecting coupling to excited states and thus the dynamics of relaxation, amply covered elsewhere.[8] Our point here was to draw the consequences of a weakly depleted condensate already existing in the ground state, not to analyze the dynamics of its formation. On the same basis, we did not consider the lifetime in the hamiltonian though of course radiative lifetime is mandatory for the effect be observed from the light field emitted by unstable polaritons. This approximation holds when the lifetime of the condensate is long as compared to the dephasing time we compute.

ACKNOWLEDGMENTS

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APPENDIX: DERIVATION OF EQ. (23)

We detail the derivation of Eq. (23) needed for the computation of $\langle S(t)\rangle$:

$$\langle n,\theta,\phi|e^{-iVt/\hbar}\exp\left[\frac{iVt}{\hbar}(a_{0\downarrow}^\dagger a_{0\downarrow}-a_{0\uparrow}^\dagger a_{0\uparrow})\right]S(0)|n',\theta,\phi\rangle$$
$$=\frac{e^{-iVt/\hbar}}{n!}\left[\langle 0,0|\sum_{\mu=0}^{n}\binom{n}{\mu}\alpha^\mu\beta^{*(n-\mu)}a_{0\uparrow}^{\mu}a_{0\downarrow}^{n-\mu}\right]$$
$$\times\exp\left[\frac{iVt}{\hbar}(a_{0\downarrow}^\dagger a_{0\downarrow}-a_{0\uparrow}^\dagger a_{0\uparrow})\right]a_{0\uparrow}a_{0\downarrow}^\dagger$$
$$\times\left[\sum_{\nu=0}^{n'}\binom{n'}{\nu}\alpha^\nu\beta^{n'-\nu}a_{0\uparrow}^{\dagger\,\nu}a_{0\downarrow}^{\dagger\,n'-\nu}|0,0\rangle\right],\tag{A1}$$

where we introduced $\alpha\equiv\cos\theta$ and $\beta\equiv e^{i\phi}\sin\theta$ as shortcuts and reverted to explicit expression (21) for $|n,\theta,\phi\rangle$. Thus, in the spin–up/down polaritons basis

$$=\frac{e^{-iVt/\hbar}}{n!}\left[\sum_{\mu=0}^{n}\binom{n}{\mu}\alpha^\mu\beta^{*(n-\mu)}\sqrt{\mu!\,(n-\mu)!}\langle n-\mu,\mu|\right]$$
$$\times\exp\left[\frac{iVt}{\hbar}(a_{0\downarrow}^\dagger a_{0\downarrow}-a_{0\uparrow}^\dagger a_{0\uparrow})\right]a_{0\uparrow}a_{0\downarrow}^\dagger$$
$$\times\left[\sum_{\nu=0}^{n'}\binom{n'}{\nu}\alpha^\nu\beta^{n'-\nu}\sqrt{\nu!\,(n'-\nu)!}|\nu,n'-\nu\rangle\right].\tag{A2}$$

We can now evaluate the operator, say, on the right expression

$$=\frac{e^{-iVt/\hbar}}{n!}\left[\sum_{\mu=0}^{n}\binom{n}{\mu}\alpha^\mu\beta^{*(n-\mu)}\sqrt{\mu!\,(n-\mu)!}\langle n-\mu,\mu|\right]$$
$$\times\left[\sum_{\nu=0}^{n'}\binom{n'}{\nu}\alpha^\nu\beta^{n'-\nu}\exp\left[\frac{iVt}{\hbar}(n'-2\nu+2)\right]\right.$$
$$\left.\times\sqrt{\nu!\,(n'-\nu)!}\sqrt{\nu(n'-\nu+1)}|\nu-1,n'-\nu+1\rangle\right].\tag{A3}$$

Since $\langle n-\mu,\mu|\nu-1,n'-\nu+1\rangle=\delta_{\mu,\nu-1}\delta_{n,n'}$, summing over ν yields

$$=\frac{e^{-iVt/\hbar}}{n!}\frac{\alpha}{\beta}\sum_{\mu=0}^{n-1}\binom{n}{\mu}\binom{n}{\mu+1}\alpha^{2\mu}|\beta|^{2(n-\mu)}$$
$$\times(\mu+1)!\,(n-\mu)!\,e^{iVt(n-2\mu)/\hbar}\tag{A4}$$

$$=e^{iVt(n-1)/\hbar}\frac{\alpha}{\beta}\sum_{\mu=0}^{n-1}\frac{n!}{\mu!\,(n-\mu-1)!}\alpha^{2\mu}|\beta|^{2(n-\mu)}e^{-2iVt\mu/\hbar}\tag{A5}$$

the sum can be computed exactly by the usual method of integration and derivation with respect to β to recover the coefficients in the binomial expansion. This way we evaluate the sum to $\beta^2(e^{-2iVt/\hbar}\alpha^2+|\beta|^2)^{n-1}n$, so that with the prefactor, the expression simplifies to

$$=n\cos\theta\sin\theta e^{i\phi}(e^{-iVt/\hbar}\cos^2\theta+e^{iVt/\hbar}\sin^2\theta)^{n-1},\tag{A6}$$

which is the expression of the text.

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