Spin noise signatures of the self-induced Larmor precession

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Bose-Einstein condensates of exciton-polaritons are known for their fascinating coherent and polarization properties. The spin state of the condensate is reflected in polarization of the exciton-polariton emission, with temporal fluctuations of this polarization being, in general, capable of reflecting quantum statistics of polaritons in the condensate. To study the polarization properties of optically trapped polariton condensates, we take advantage of the spin noise spectroscopy technique. The ratio between the noise of ellipticity of the condensate emission and its polarization plane rotation noise is found to be dependent, in a nontrivial way, on the intensity of CW nonresonant laser pumping. We show that the interplay between the ellipticity and the rotation noise can be explained in terms of the competition between the self-induced Larmor precession of the condensate pseudospin and the static polarization anisotropy of the microcavity.

Introduction. The bosonic condensates of exciton-polaritons are unique physical objects exhibiting spontaneous coherence in hybrid driven-dissipative systems. One of their most fascinating properties is an ability to emit a coherent light in a way typical to the conventional lasers but with no need of any population inversion [1]. The exciton polaritons arise due to the strong coupling between the semiconductor excitons and the quantized light field [2]. Therefore the macroscopic coherent state of the polariton condensate inherits the spin degree of freedom from both polariton constituents, which is correlated with the polarization of the light emitted by the condensate. In particular, polaritons formed by the coupling of the heavy hole exciton with the photon, have two possible spin projections on the structure growth axis (either +1 or −1) which correspond to the right and left circular polarization of the emitted photons [3].

Although the exciton-polaritons have been detected in a wide variety of systems [4–8] including Fabry–Perot microcavities [9–11] and planar waveguides [12], the most thoroughly investigated system is a semiconductor microcavity. Polaritons can be excited optically both resonantly and nonresonantly [13, 14], as well as by the electric current even at room temperature [4, 15, 16].

Despite the well developed theoretical basis of exciton-polariton emitters, there is still a number of experimentally unrevealed aspects of their microscopic dynamics. In particular, the effect of self-induced Larmor precession, well known theoretically [17, 18] and studied in pulsed and CW resonant [19] experiments, was never observed under nonresonant excitation of the condensate. This effect consists in the precession of the Stokes vector of the condensate of exciton-polaritons about an effective magnetic field induced by the condensate itself and oriented along the structure axis. The effect is a peculiar manifestation of the anisotropy of the exchange interaction between excitons in semiconductor quantum wells. In this context, the spin noise spectroscopy (SNS) technique, widely used in atomic and solid state systems for spin dynamics investigation [20–22], was found to be extremely helpful in exciton-polariton condensate studies [23], being informative even in the case when the information seems to be totally lost [24].

In this paper, we employ the SNS to study the noisy polarization properties of the optically trapped polariton condensate. Quite surprisingly, even at almost fully non-polarized condensate emission, the behaviour of the emission noise demonstrates a strong asymmetry between the ellipticity noise and the polarization plane azimuth noise. We attribute the observed asymmetry to the presence of the self-induced Larmor precession of polarization of the continuously pumped polariton condensate.

This observation paves the way towards application of semiconductor microcavities in optical spintronic devices as proposed in [25].

Experimental setup. To be specific, we study the polarization properties of the polariton condensate formed in the ultra-high-finesse ($Q \approx 31\,000$) that corresponds...
to the polariton lifetime $\tau_p \approx 100 \text{ ps}$) 5λ/2 microcavity. The cavity is formed by $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ gap with four sets of three 12 nm GaAs QWs located at the antinodes of the cavity mode. Top (bottom) Bragg mirror is comprised of 45 (50) AlAs/$\text{Al}_{0.15}\text{Ga}_{0.85}\text{As}$ layers.

The polariton condensate is excited by the nonresonant pump of continuous-wave Ti:Sapphire T&D-scan laser (1), see Fig. 1. The wavelength of the pump was chosen to match the first next to stop-band reflectivity dip at $\sim 766 \text{ nm}$. The pump laser emission was directed to a FullHD MEMS matrix Texas Instruments DLP Lightcrafter 6500 (2) which serves for the manipulation of the pump beam spatial profile. The reflected monochromatic image was collimated by a 15 cm lens (3) and transmitted through a nonpolarizing beamsplitter to Mitutoyo 50× APO NIR microobjective (4) to the sample (5) that was placed in a closed-cycle cold-finger cryostat Montana Cryostation. The emitted light passed in an autocollimation geometry through an optical low-pass filter (6) to a conventional spin noise detection setup [21, 23], which consists of a quarter-wave plate (7), a polarizing beam splitter (8), the balanced photoreceiver NewFocus 2107 (9) and the spectrum analyzer Tektronix RSA5103A (10). The real-space distribution of the condensate emission was detected by means of a nonpolarizing beam sampling plate (11) and the infrared camera Andor Luca R-604 (12).

Polarisation measurements require a special care about the spatial and spectral homogeneity of the condensate state. An undesirable fragmentation of the condensate suppresses the polarisation signal collected from the whole condensate. In our case, a high degree of homogeneity of the QW interface along with the long polariton lifetime allows for the trapping of the condensate with the pure shape. Applying the specific spatial profile of the nonresonant pump spot (see the insets in Fig. 2) with the DLP modulator we confine polaritons in an optical trap. The pump creates a cloud of incoherent excitons [26] which push polaritons to the non-excited region due to the exciton repulsion. The polariton confinement leads to the formation of a more pure condensate state than in the case of a Gaussian pump spot profile.

The measurements of the polarization degree of the polariton emission (PE) have shown that the condensate was essentially unpolarized at the most of the applied pump beam profiles. Regarding the implementation of the SNS technique, it guarantees that the photocurrent balance will be kept at any orientation angle of the spin-noise detection scheme. However the control of the mutual orientation of quarter-wave plate fast axis and the polarisation beam splitter main axis (characterized by angle $\theta$) allows to switch between the detection of the polarisation plane azimuth rotation noise, which we call hereafter the rotation noise (RN), and the ellipticity noise (EN), at $\theta = 0$ and $\theta = 45^\circ$, respectively.

The measured polarization noise parameters can be related to the Stokes parameters of the polariton condensate emission. The latter, in turn, directly correlates with the condensate pseudospin vector $\mathbf{S} = 1/2(\mathbf{\Psi}^\dagger \sigma \mathbf{\Psi})$, where $\sigma_{x,y,z}$ are the Pauli matrices and $\mathbf{\Psi} = (\Psi_+, \Psi_-)$ is the condensate order parameter whose components define two possible projections of the polariton spin on the structure growth axis, see Fig. 1b. In particular, we take the horizontal $\leftrightarrow$ and the vertical $\uparrow \downarrow$ Stokes polarization (polariton pseudospin) axes coincident with the Cartesian axes of the PBS. Then, at $\theta = 0$ the spin noise detection scheme tracks the noisy dynamics of the $S_x$ component of the polariton pseudospin, see Fig. 1b. Likewise, if the quarter-wave plate is tilted at $\theta = 45^\circ$, the circularly polarized component of emission is converted to the linear polarization, and the balanced scheme detects the intensity variations of circularly polarized components of the emitted light, i.e. the fluctuations of $S_y$ parameter.

Fixing the shape of the pump spot, we performed several tens of pump intensity scans for both $\theta = 0$ and $\theta = 45^\circ$ quarter-wave plate positions. For each value of the pump intensity we collected the integral of the balanced photodiode current noise power in the frequency range of [0...1] MHz. Although the obtained dependencies (see Fig. 2) varied for different excitation beam profiles, in a vast majority of cases, especially for the highly symmetrical pumps, the noise behaviour follows the same typical scenario. In particular, the rotation noise dominates over the noise of ellipticity at the weak pump, typically below lasing threshold (see the insets of Fig. 2). With the increase of the pump intensity, the noise signals of both types progressively grow. However, the EN power growth rate typically exceeds the increment of the RN power. As a result, at some point above the polariton lasing threshold EN-signal becomes greater than the RN.

**Discussion** We interpret the observed behavior of the condensate polarization noise in terms of the interplay between the static polarization anisotropy, which tends to suppress the circular polarization, and the spin-dependent polariton-polariton interactions, which trigger the self-induced Larmor precession of the condensate pseudospin. We describe the state of the fluctuating order parameter of the polariton condensate in the presence of the non-resonant pump by the stochastic driven-
dissipative Gross-Pitaevskii equation [27]:

\[ i\hbar \partial_t \Psi_\pm = \left( \alpha_1 |\Psi_\pm|^2 + \alpha_2 |\Psi_\mp|^2 + \alpha_\nu n \right) \Psi_\pm + \frac{i\hbar}{2} \left( Rn - \gamma_\nu \right) \Psi_\pm + \frac{\delta_\nu}{2} \Psi_\mp \mp \sqrt{D} \eta_\nu(t), \]  

(1)

for the two-component complex order parameter \( \Psi = (\Psi_+, \Psi_-) \), whose indices correspond to polariton \( \pm \) spin projections on the structure growth axis. Coefficients \( \alpha_1, \alpha_2 \) define the interaction strengths between polaritons with the same and the opposite spins, respectively, while \( \alpha_\nu \) stands for the interaction of the polaritons from the condensate with the incoherent excitons created by the nonresonant pump. \( \gamma_\nu \) is the polariton lifetime. The quantity \( \delta_\nu \) in the last but one term describes the splitting of the linear polarizations that stems from the optical anisotropy of the microcavity and, in our case, is ascribed to the inevitable mechanical strain of the structure due to the lattice disregistry of the Bragg mirror layers.

The random fluctuations of the order parameter are accounted for by the last term in Eq. (1). Taking into account the high homogeneity of the sample microcavity structure and the temporal stability of the pumping laser, we attribute the main source of these fluctuations to the scattering from the incoherent reservoir to the condensate state. Each act of the scattering perturbs the phase of both spin components of the condensate, thus randomizing its polarization. We simulate \( \eta_\nu(t) \) as a Wiener process with the spectral density \( D = \langle \eta_i(t) \eta_j(t') \rangle = 1/2 Rn \delta(t-t') \delta_{ij} \), - see [27], where \( i, j = +, - \), \( n \) corresponds to the incoherent exciton density and \( R \) is the scattering rate. Thus the magnitude of the order parameter fluctuations appears to be dependent on the pump power.

Since the pumping laser frequency was tuned far above the exciton reservoir, we assume the loss of the memory of the pumping laser polarization, considering the spin-unpolarized reservoir, which integrated density \( n \) is governed by the rate equation

\[ \partial_t n = P - \gamma_r n - Rn \left( |\Psi_+|^2 + |\Psi_-|^2 \right), \]  

(2)

where \( P \) defines the pump intensity and \( \gamma_r \) is a reservoir population relaxation parameter.

Note that for simplicity we neglect by the spatial distributions of \( \Psi \) and \( n \). This approach allows us to describe the typical behaviour of the PE noise which is independent on the optical trap shape.

Eqs. (1) and (2) were solved numerically on the timescales of several tens of nanoseconds. The polarization noise properties are governed by the behaviour of the pseudospin (Stokes) vector of the condensate, whose components in the circular polarization basis read \( S_x = \text{Re}(\Psi^* \Psi_+), S_y = \text{Im}(\Psi^* \Psi_+), S_z = (|\Psi_+|^2 - |\Psi_-|^2)/2 \). The results of our simulations are shown in Fig. 3. The stochastic dynamics predicts the width of the fluctuation spectrum to be comparable to the spectral line width, that is of tenth of THz, see Fig. 3a,d. The bandwidth of experimentally used balanced photodiode was much smaller (limited by 1 MHz). Therefore the noise power measured with the balanced photoreceiver of the SNS scheme corresponds to the calculated spectral density \( Q_\beta (\omega) = \int B_\beta(\tau) e^{-i\omega \tau} d\tau \) taken at zero frequency, \( \omega = 0 \), that is \( Q_\beta (0) = \int B_\beta(\tau) d\tau \). Here

\[ B_\beta(\tau) = T^{-1} \int_0^T S_\beta(t) S_\beta(t + \tau) dt \]  

is a correlation function of the quantity \( S_\beta \) with \( \beta \) standing for the indices \( (x, y, z) \) and \( T \) being the time of calculations.

The calculated pump-intensity-dependence of the noise power, Fig. 3c, agrees with the experimentally observed asymmetry between the subthreshold and the above-threshold regimes of the polarization noise behaviour.

The observed asymmetry can be understood examining the conservative dynamics of the Stokes vector, \( \partial_t S \propto -\Omega \times S \), which corresponds to its precession about the effective magnetic field \( \Omega = (\delta_l, 0, (\alpha_1 - \alpha_2) S_z) \). The \( x \)-component of this field, \( \Omega_x = \delta_l \), arises from the splitting of linear polarizations and accounts for the rotation of the condensate pseudospin \( S \) about \( x \) axis. The field strength \( \Omega_x \) does not depend on the pumping level, i.e. this precession occurs with the same frequency both below and above the threshold. In contrast, the \( z \)-dependent effective field \( \Omega_z = (\alpha_1 - \alpha_2) S_z \) does depend on the pump and appears to be suppressed at the small condensate occupancies, \( S_z \rightarrow 0 \), typically below the threshold. An example of the fluctuation dynamics of the condensate pseudospin at the subthreshold pump is shown in Fig. 3b. Due to the precession of the pseudospin \( S_y \) and \( S_z \) components their zero-frequency noise power turns out to be suppressed, causing the RN power \( Q_x(0) \) to exceed the EN power \( Q_z(0) \), see Fig. 3c and [28].
An increase of the pump power above the threshold results in the growth of the condensate occupancy and the increase of the effective magnetic field generated due to spin-dependent polariton-polariton interactions. Due to the strong spin anisotropy of the polariton-polariton interactions, which are much stronger for the same spin than for the opposite spin polaritons ($\alpha_1 \neq \alpha_2$), the fluctuations of the population imbalance between the circularly polarized condensate components $S_z$ trigger the pseudospin rotation about $z$ axis. This effect is known as the pseudospin self-induced Larmor precession [17].

Note that far above the threshold the fluctuations of the order parameter which are governed by the reservoir occupancy become so strong that the pseudospin vector spans all the Poincaré sphere, see. Fig. 3e. Therefore the degree of polarization of the condensate emission approaches zero that is consistent with our experimental results.

Although the time integrated value of $S_z$ is zero, the amplitude of its fluctuations grow with the pump power increase. Therefore, above the threshold the time averaged modulus of $\Omega_z$ grows up and at the certain pumping intensity, which is typically close to the threshold, becomes larger than $\Omega_z$. The effective magnetic field vector tilts to $z$ axis. As a result the fast oscillations of the instant values of $S_x$ and $S_y$ components, being averaged over time that essentially exceeds the inverse effective field strength, lead to suppression of RN power compared to the fluctuations of $S_z$.

This approach allows reproducing of the characteristic behaviour which is qualitatively the same for any pump beam profile. An influence of the specific shape of the pump beam can be accounted for by rescaling of the reservoir to condensate scattering rate $R \rightarrow \beta R$. The factor $\beta < 1$ accounts for the spatial overlap between the condensate and the reservoir, which is dependent on the pump beam profile and is expected to decrease with the growth of the size of the optical trap. Our simulations show that the variation of $\beta$ alters the position of the crossing point of the EN and RN pump power dependencies.

In conclusion, we show that the SNS technique is capable to reveal the intrinsic fluctuating dynamics of continuously pumped exciton-polariton system even if the condensate emission is almost completely unpolarized. We demonstrate that the stochastic polarization dynamics is a key reason of the vanishing degree of polarization of the trapped condensate. The behaviour of the polarization noise, described in terms of the ellipticity noise and the polarization plane azimuth noise, demonstrates a strong asymmetry between the subthreshold and above-threshold regimes. This asymmetry stems from the competition of the linear polarization splitting and the effect of the self-induced Larmor precession of the condensate pseudospin. In particular, we found that below the polariton lasing threshold the fluctuation dynamics is superimposed by the pseudospin precession about the effective magnetic field oriented in the equatorial plane of the Poincaré sphere. We ascribe the appearance of this effective magnetic field to the microcavity mirror birefringence which arises from the mechanical strain caused by the lattice mismatch of the materials. Above the conden-
sation threshold the effect self-induced Larmor precession builds up as the population of circular polarization states grows up. These concurring effects lead to the intersection of the EN and RN pump-power-dependencies.

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[1] A. Imamo˘ glu, R. J. Ram, S. Pau and Y. Yamamoto, Phys. Rev. A. 53, 4250–4253 (1996).
[2] A. Kavokin and G. Malpuech, Cavity Polaritons, edited by Y. M. Agranovich (Elsevier, North Holland, Amsterdam, 2003).
[3] I. A. Shelykh, Yuri G. Rubo, G. Malpuech, D. D. Solnyshkov, and A. Kavokin, Phys. Rev. Lett. 97, 066402 (2006).
[4] Christmann, G., Butté, R., Feltin, E., Carlin, J. F. and Grandjean, N., Appl. Phys. Lett. 93, 51101 (2008).
[5] Feng Li, L. Orosz, O. Kamoun, S. Bouchoule, C. Brimont, P. Disseix, T. Guillet, X. Ladosse, M. Leroux, J. Leymarie, M. Mexis, M. Mihailovic, G. Patriarche, F. Réveret, D. Solnyshkov, J. Zuniga-Perez, and G. Malpuech, Phys. Rev. Lett. 110, 196406 (2013).
[6] C. P. Dietrich et al., Sci. Adv. 2, e1600666 (2016).
[7] S. Pirotta et al., Appl. Phys. Lett. 104, 051111 (2014).
[8] T. Ellenbogen and K. B. Crozier, Phys. Rev. B 84, 161304 (2011).
[9] D. Bajoni, P. Senellart, E. Wertz, I. Sagnes, A. Miard, A. Lemaître, and J. Bloch, Phys. Rev. Lett. 100, 047401 (2008).
[10] A. Das, P. Bhattacharya, J. Heo, A. Banerjee, and W. Guo, Proc. Natl. Acad. Sci. USA 110, 2735 (2013).
[11] S. Christopoulos, G. B. H. von Högersthal, A. Grundy, P. G. Lagoudakis, A. V. Kavokin, J. J. Baumberg, G. Christmann, R. Butté, E. Feltin, J.-F. Carlin, and N. Grandjean, Phys. Rev. Lett. 98, 126405 (2007).
[12] Jamadi, O., Reveret, F., Disseix, P. et al., Light Sci. Appl. 7, 82 (2018).
[13] Wertz E., Ferrier L., Solnyshkov D. D. et al., Appl. Phys. Lett. 95 051108 (2009).
[14] P. Tsotsis, P. S. Eldridge, T. Gao, S. I. Tsintzos, Z. Hatzopoulos, and P. G. Savvidis, New J. Phys. 14, 023060 (2012).
[15] S. Christopoulos et al., Phys. Rev. Lett. 98, 126405 (2007).
[16] P. Bhattacharya et al., Phys. Rev. Lett. 112, 236802 (2014).
[17] I. Shelykh, G. Malpuech, K. V. Kavokin, A. V. Kavokin, and P. Bigenwald, Phys. Rev. B 70, 115301 (2004).
[18] F. P. Laussy, I. A. Shelykh, G. Malpuech, and A. V. Kavokin, Phys. Rev. B 73, 035315 (2006).
[19] D. N. Krizhanovskii, D. Sanvitto, I. A. Shelykh, M. M. Glazov, G. Malpuech, D. D. Solnyshkov, A. Kavokin, S. Ceccarelli, M. S. Skolnick, and J. S. Roberts, Phys. Rev. B 73, 073303 (2006).
[20] M. Müller, M. Oestreich, M. Römer, and J. Hübner, Physica E, 43, 569 (2010).
[21] V. S. Zapasskii, Adv. Opt. Photonics 5, 131 (2013).
[22] J. Hübner, F. Berski, R. Dahbashi, and M. Oestreich, Physica Status Solidi B 251, 1824 (2014)
[23] I. I. Ryzhov, M. M. Glazov, A. V. Kavokin, G. G. Kozlov, M. Assmann, P. Tsotsis, Z. Hatzopoulos, P. G. Savvidis, M. Bayer, and V. S. Zapasskii, Phys. Rev. B 93, 241307(R) (2016).
[24] G. G. Kozlov, I. I. Ryzhov, A. Tzimis, Z. Hatzopoulos, P. G. Savvidis, A. V. Kavokin, M. Bayer, and V. S. Zapasskii, Phys. Rev. A 98, 043810 (2018).
[25] I. Shelykh, K. V. Kavokin, A. V. Kavokin, G. Malpuech, P. Bigenwald, H. Deng, G. Weihs, and Y. Yamamoto, Phys. Rev. B 70, 035320 (2004).
[26] A. Askitopoulos, O. Ohadi, A. V. Kavokin, Z. Hatzopoulos, P. G. Savvidis, and P. G. Lagoudakis, Phys. Rev. B 88, 041308(R) (2013).
[27] D. Read, T. C. H. Liew, Y. G. Rubo, and A. V. Kavokin, Phys. Rev. B 80, 195309 (2009).
[28] M. M. Glazov, M. A. Semina, E. Ya. Sherman, and A. V. Kavokin, Phys. Rev. B 88, 041309 (2013).