Giant spin Meissner effect in a nonequilibrium exciton-polariton gas

M. Król,[1,*] R. Mirek,[1] D. Stephan,[1] K. Lekenta,[1] J.-G. Rousset,[1] W. Pacuski,[1] A. V. Kavokin,[2,3,4] M. Matuszewski,[5] J. Szczytko,[1] and B. Piętka[1,†]

[1]*Institute of Experimental Physics, Faculty of Physics, University of Warsaw, ulica Pasteura 5, PL-02-093 Warsaw, Poland*
[2]*Institute of Natural Sciences, Westlake University, No. 18, Shilongshan Road, Cloud Town, Xihu District, Hangzhou, China*
[3]*National University of Science and Technology "MISIS," 4, Leninskiy prospekt, 119049 Moscow, Russia*
[4]*Spin Optics Laboratory, St. Petersburg State University, Ul'anovskaya 1, Peterhof, St. Petersburg 198504, Russia*
[5]*Institute of Physics, Polish Academy of Sciences, aleja Lotników 32/46, PL-02-668 Warsaw, Poland*

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The suppression of Zeeman energy splitting due to spin-dependent interactions (the spin Meissner effect) was predicted to occur within a Bose-Einstein condensate. We report a clear observation of this effect in semimagnetic microcavities which exhibit a giant Zeeman energy splitting between two spin-polarized polariton states as high as 2 meV and demonstrate that a partial suppression of the energy difference occurs already in the uncondensed phase in a striking similarity to the behavior of up-critical superconductors in the fluctuation-dominated regime. These observations are explained quantitatively by a kinetic model accounting for both the condensed and uncondensed polaritons and taking into account the nonequilibrium character of the system.

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I. INTRODUCTION

One of the defining properties of a superconductor is the expulsion of external magnetic field from its interior by surface currents appearing on the boundary. This phenomenon, known as the Meissner effect [1], has its analog in a neutral bosonic condensate with spin. In this case, the minimization of free energy in the presence of magnetic field leads to a complete screening of the Zeeman splitting by the interactions between two spin subsystems [2]. Indeed, the imbalance between populations of spin components results in an effective magnetic field that exactly cancels the external magnetic field in the ground state of the system. This exact compensation of the external magnetic field effect is expected to occur at fields up to some critical value dependent on the g factor of bosonic quasiparticles, the interaction constants of spin-parallel and spin-antiparallel bosons, and the concentration of bosons.

Exciton-polaritons in semiconductor microcavities constitute an example of spinor bosonic quasiparticles: a mixture of matter excitation in semiconductor quantum wells (excitons) and photons confined in a cavity structure [3,4]. The excitons that can couple with light have a ± 1 spin degeneracy and therefore couple, respectively, with photons of opposite chirality $\sigma^{\pm}$, forming two subsystems of exciton-polaritons of opposite spins. They can be considered bosonic quasiparticles with a $1/2$ pseudospin. Most importantly, the interactions between polaritons are spin dependent [5–9]. Typically, polaritons with the same spin projection on the quantum well axis strongly repel, while polaritons with opposite spin projections weakly attract [10,11]. This interaction constant difference together with population imbalance is responsible for the appearance of the spin Meissner effect in a polariton condensate. On the one hand, due to the Zeeman effect and thermalization processes, polaritons tend to orient their spins in external magnetic fields; on the other hand, polaritons try to minimize the free energy due to the strong repulsion of polaritons with the same spin. Therefore, there is an interplay between spin polarization induced by external magnetic field and polariton-polariton interaction, which leads to suppression of spin splitting, which acts effectively as the expulsion of magnetic field from the superconductor interior.

The theoretical prediction of the spin Meissner effect in a polariton condensate presented in Ref. [2] was followed by multiple experimental works [12–16]. Suppression of the Zeeman splitting [12], even up to sign reversal [14], was reported but was accompanied by an unexpected linear polarization behavior and deviations from the predictions of the thermal equilibrium model in the polarization splitting dependence on the field [17,18]. Most authors explain deviations from the theory as being due to the nonequilibrium polariton dynamics [13–15], while the stationary regime close to thermal equilibrium in polariton condensates is achieved only in the last generation of samples [19,20]. In electrically driven polariton lasers, suppression of the Zeeman splitting was observed in the polariton-lasing regime [21], but in another realization of a similar experiment a circular polarization of the emission was attributed to the Zeeman splitting [22].

The most important parameter that up to now has made the proper experimental observation of those effects difficult is the small energy splitting of exciton-polaritons of opposite spins (weak Zeeman effect). Using semimagnetic semiconductors allows us to overcome this limitation. Semimagnetic materials were also considered in theoretical works investigating the spin Meissner effect [23]. The theory assuming the minimization of free energy of the condensate energy for a fixed position of magnetic ions in the system predicted a

*mateusz.krol@fuw.edu.pl
†barbara.pietka@fuw.edu.pl

linear polarization of a polariton condensate in equilibrium and an increasing degree of circular polarization with increasing magnetic field. Our first results in CdMnTe-based semimagnetic semiconductor microcavities showed an increase in the degree of circular polarization of the condensate with magnetic field [24,25] but did not show strong evidence of quenching of the Zeeman splitting. Here, we demonstrate that in a specially designed system supporting a semimagnetic polariton condensate with giant spin splitting [26] and a careful choice of excitation parameters suppression of the Zeeman splitting around the condensation threshold is observed, which we believe is a clear signature of the spin Meissner effect predicted in Ref. [2]. The magnitude of the quenching is as high as 2 meV at 6 T. We extend also the theoretical model provided there to account for a nonequilibrium character of the polariton condensate.

Moreover, we present experimental evidence of a new regime of a polariton gas, which we call the reservoir-dominated regime, in analogy to the fluctuation-dominated regime of fluctuating superconductors. The fluctuation-dominated regime is characterized by the occurrence of the partial Meissner effect [27]. We observe an analogous phenomenon in a polariton gas, with a partial spin Meissner effect below the laser threshold. This is a very different observation compared to previous works where the features of the spin Meissner effect were detected only above the polariton lasing threshold. The observation of this regime is of fundamental importance as it shows the similarity of our bosonic system and fluctuating superconductors [27]. The reservoir-dominated phase appears in our system as it offers a supplementary mechanism of fluctuations: the magnetization fluctuations in the semimagnetic structure that affect the potential seen by exciton-polaritons. This specific feature of a semimagnetic cavity makes it an excellent test bed for revealing fluctuation effects in a bosonic polariton gas.

II. EXCITON-POLARITON CONDENSATE IN MAGNETIC FIELD

We investigated a nonmagnetic CdTe-based microcavity with a quantum well doped with magnetic Mn ions, presented schematically in Fig. 1(a) and described in more detail in [28,29]. In the CdMgZnTe microcavity structure and CdMnTe semimagnetic material of the quantum well, the *s*, *p*-*d* exchange interaction between the localized *d*-shell electrons of the magnetic ions and the *s*-shell electrons and *p*-shell holes leads to magneto-optical effects such as giant Faraday rotation and giant Zeeman splitting [30]. The scheme of the structure is illustrated in Fig. 1(a). In Ref. [26] we demonstrated a giant Zeeman splitting of exciton-polaritons in the same microcavity structure, where magnetic ions are present only in quantum wells, affecting only the excitonic component of the polariton state. We have shown that the external magnetic field can induce condensation by reducing the condensation threshold power due to the decrease of the available density of states up to a factor of 2, which reduces the condensation threshold [24]. Moreover, we demonstrated the creation of a spin multicomponent condensate and the possibility to tune it smoothly to a single-component condensate with an external parameter, i.e., excitation power and/or magnetic field [25].

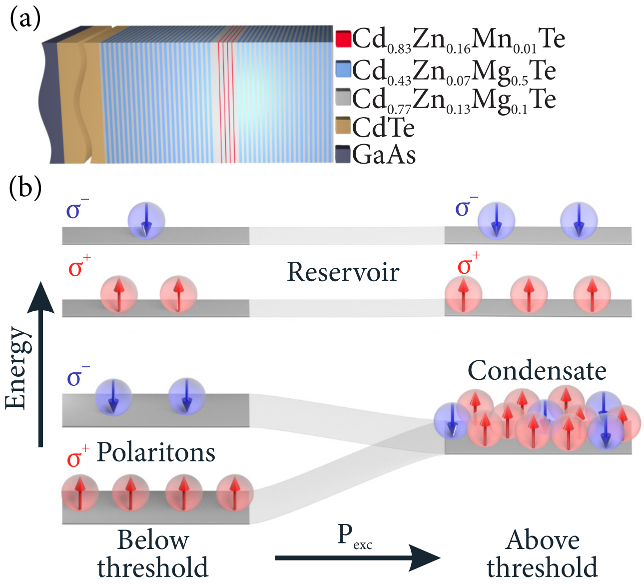

FIG. 1. (a) Scheme of the investigated microcavity with 20 (22) top (bottom) CdZnMgTe DBR pairs and four quantum wells doped with magnetic Mn^{2+} ions. (b) Energy levels in external magnetic field with increasing excitation power. Above the threshold the condensate Zeeman splitting is suppressed, while the reservoir splitting is still present.

The results presented here demonstrate a different approach to the spin Meissner effect of semimagnetic exciton-polariton spinor condensates, where the magnetic interactions play a crucial role. We create a polariton condensate at several values of the external magnetic field by increasing the excitation power, which is schematically shown in Fig. 1(b). At fixed magnetic field and below the condensation threshold [Fig. 1(b)] the system exhibits giant Zeeman energy splittings both in the polariton state (along the lower polariton branch) and in the excitonic reservoir (which can be traced at high emission angles, where polaritons are mostly excitonic). With increasing pumping power a quenching of the Zeeman splitting at the polariton mode followed by the formation of polariton condensate is observed. The uncondensed polaritons that form the reservoir still exhibit a giant Zeeman splitting. Measurements were performed for negative exciton-photon detuning of −12 meV with Rabi splitting equal to 7.7 meV without the external magnetic field. Figure 2 shows the evolution of the experimental emission spectra of semimagnetic polaritons at a magnetic field of 4 T with increasing excitation power. The spectra for other values of magnetic field ranging from 0 to 6 T are provided in the Supplemental Material [31]. As the excitation power increases, we observe the polariton condensation at the bottom of the lower polariton branch that is accompanied by a quenching of the energy difference between the counterpolarized signals (in σ^+ and σ^- polarization detection). Uncondensed polaritons, described within a two-mode coupling model [32] and marked by solid and dashed lines in Fig. 2, show Zeeman energy splitting, as anticipated in the linear regime [26,33]. The polariton condensation at the bottom of the lower polariton branch is also revealed by a nonlinear increase in the emission intensity, linewidth narrowing, and energy blueshift due to the interactions present

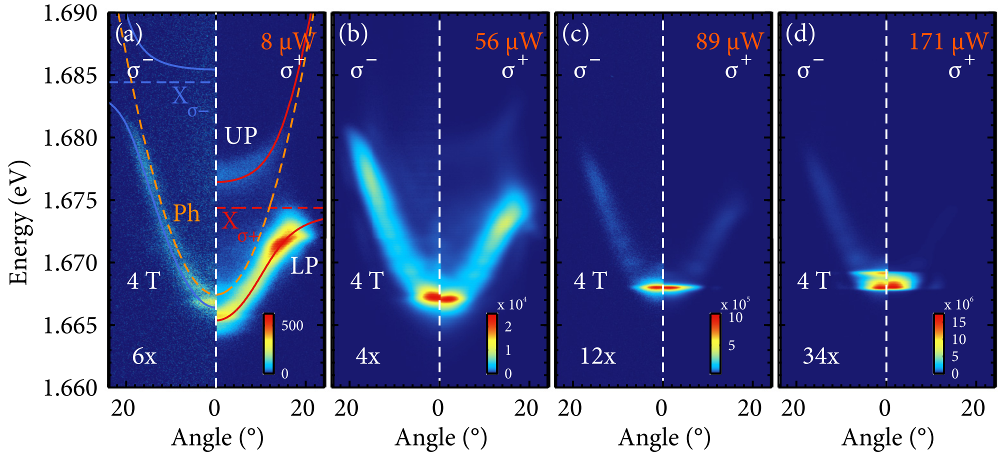

FIG. 2. Experimental data illustrating the semimagnetic condensate formation upon increasing excitation power at 4 T. The system is excited with a linearly polarized picosecond-pulsed laser at the energy of the first Bragg minimum of the structure at the high-energy side (approximately 1.746 eV). The photoluminescence spectra are time integrated and are angularly resolved with the angle corresponding to the polariton in-plane momentum. The dashed lines represent the energy modes of the uncoupled exciton and photon system, while polariton modes are marked by red and blue solid lines, corresponding to σ^+ and σ^- polarization detections, respectively.

in the system, which is illustrated in detail in the Supplemental Material [31]. The condensation threshold can be found for each magnetic field, and it equals 60 μW at 0 T. It slightly decreases in magnetic field, in agreement with the previous studies [24].

In Figs. 3(a)–3(c) we plot the energy of the emission lines as a function of the excitation power for both predominantly σ^+ and σ^- polarized states. We observe that with an increase in the excitation power the energy difference between σ^+ and σ^- states decreases; moreover, at the threshold the energy difference becomes smaller than the linewidth and cannot be resolved. With a further increase of the excitation power we observe that both spin components of the condensate have the same energy, which increases due to the interactions in the system. Figure 4(a) illustrates the energy difference between the two circularly polarized components of the condensate, and Fig. 4(b) shows the accompanying splitting of uncondensed polaritons that represents the Zeeman splitting of the excitonic reservoir. The threshold power for each magnetic field is marked by the crosses. We believe that reduction

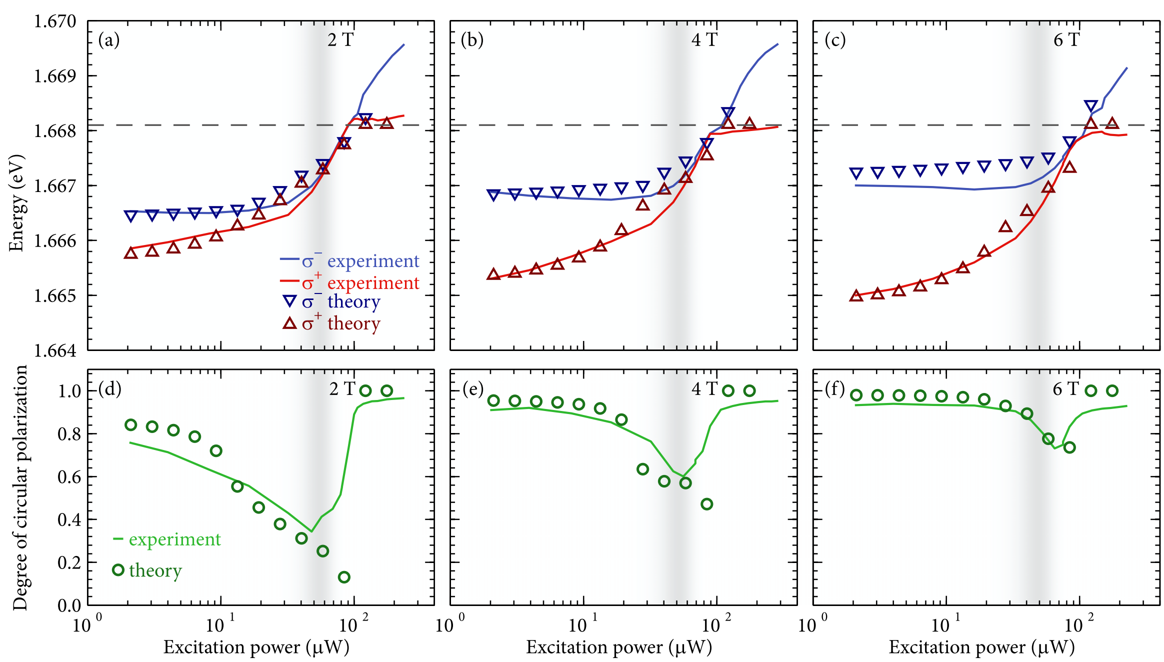

FIG. 3. (a)–(c) The energy and (d)–(f) degree of circular polarization of the signal at the bottom of lower polariton branch versus excitation power in magnetic fields of 2, 4, and 6 T. The energy difference between σ^+ and σ^- polariton states decreases with the pumping power, with complete suppression of the splitting close to the condensation threshold, where the minimum of the DOCP occurs. Experimental results are marked with solid lines, with the fitted model represented with points. The condensation threshold, marked by the shading, is 60 μW at 0 T and is slightly decreasing in magnetic field.

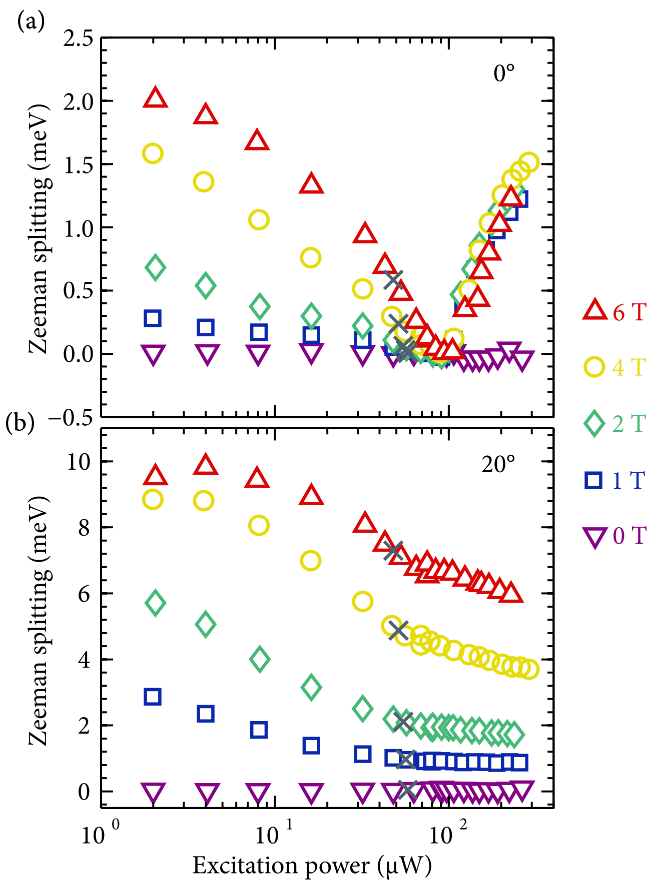

FIG. 4. Energy splitting between two spin components of (a) the exciton-polariton condensate at the bottom of the lower polariton branch and (b) the noncondensed polariton reservoir detected at a 20° angle. The condensation threshold power at each magnetic field is marked with a gray cross.

of the Zeeman splitting of the condensate down to zero is a clear signature of the spin Meissner effect. It shows the importance of spin-dependent interactions within the condensate. In addition, a partial reduction of Zeeman splitting with increasing pumping power is observed for a polariton gas well below the condensation threshold, a behavior which is not predicted by existing models.

III. THEORETICAL MODEL

To explain the above results, indicating that reduction of giant Zeeman splitting can be clearly recognized even below the condensation threshold, which is visible in Fig. 4(a), we introduce a phenomenological model of the polariton reservoir and condensate dynamics. Polariton kinetics are accounted for in a way similar to that in several previous works [34–36], where spinor polariton condensates were investigated. Additionally, we describe the spin Meissner effect by adapting the description formulated in Refs. [2,37] but generalized to the system in a nonequilibrium state. To account for both the reservoir dynamics and condensate formation, as well as the photon lasing at high pumping powers, we formulate the equations for the reservoir density $n_{\mathrm{R}}(t)$ and its average spin $\mathbf{S}_{\mathrm{R}}(t)$, the condensate density $n_{\mathrm{c}}(t)$, assuming that the spin of the condensate always corresponds to the minimum of energy $\mathbf{S}_{\mathrm{c}}(t)=\mathbf{S}_{\mathrm{c}}^{\min}(t)$, and the lasing mode occupation $n_{\mathrm{L}}(t)$,

$$\frac{\partial n_{\mathrm{R}}}{\partial t}=P-\gamma_{\mathrm{R}}n_{\mathrm{R}}-R_{\mathrm{sc}}n_{\mathrm{R}}n_{\mathrm{c}}-R_{\mathrm{L}}n_{\mathrm{R}}n_{\mathrm{L}},$$
$$\frac{\partial \mathbf{S}_{\mathrm{R}}}{\partial t}=-\gamma_{\mathrm{rel}}\left(\mathbf{S}_{\mathrm{R}}-\mathbf{S}_{\mathrm{R}}^{\min}\right)-R_{\mathrm{sc}}n_{\mathrm{c}}\mathbf{S}_{\mathrm{R}}-\gamma_{\mathrm{R}}\mathbf{S}_{\mathrm{R}},$$
$$\frac{\partial n_{\mathrm{c}}}{\partial t}=R_{\mathrm{sc}}n_{\mathrm{R}}n_{\mathrm{c}}-\gamma_{\mathrm{c}}n_{\mathrm{c}},\qquad \frac{\partial n_{\mathrm{L}}}{\partial t}=R_{\mathrm{L}}n_{\mathrm{R}}n_{\mathrm{L}}-\gamma_{\mathrm{L}}n_{\mathrm{L}},$$

where P is the pumping rate; $\gamma_{\mathrm{R,c}}$ are excitonic and photonic decay rates; $R_{\mathrm{sc}}\approx R_0+R_1E_c$ is the reservoir-condensate scattering rate, which is approximately linearly dependent on the condensate energy; R_{L} is the rate of scattering into the lasing mode; and $\gamma_{\mathrm{rel}}=\gamma_{\mathrm{phonon}}+\Gamma_{\mathrm{int}}n_{\mathrm{R}}$ is the reservoir relaxation rate, which accounts for the phonon-mediated relaxation γ_{phonon} and interaction-mediated relaxation Γ_{int}. The reservoir and condensate spins that minimize the energy of the system $\mathbf{S}_{\mathrm{R,c}}^{\min}$ are calculated as in [2,37], from the minimization of the free energy

$$F=-2\Omega_zS^z+2\Omega_xS^x+\frac{\alpha}{2}(n_+^2+n_-^2)-\mu n, \tag{1}$$

where Ω_z is the excitonic giant Zeeman splitting appearing due to the magnetic field parallel to the sample growth axis; Ω_x is the linear polarization splitting due to the sample anisotropy or the residual transverse magnetic field; $S^{x,z}=S_R^{x,z}+XS_c^{x,z}$ is the z component of the total excitonic spin, where X is the Hopfield coefficient of condensed polaritons; $n^{\pm}=n_{\mathrm{R}}^{\pm}+Xn_{\mathrm{c}}^{\pm}$, where $n_{\mathrm{R,c}}^{\pm}=(n_{\mathrm{R,c}}/2)\pm S_{\mathrm{R,c}}^z$; α is the parallel spin interaction coefficient; and we neglect the interaction between polaritons with antiparallel z-spin projection. Theoretical and experimental estimates indicate that such interaction is much weaker than in the parallel spin configuration [38], and we found that the results presented in our work can be well explained by neglecting polariton-polariton interactions in the antiparallel spin configuration.

IV. REDUCTION OF POLARITON ZEEMAN SPLITTING BELOW THE CONDENSATION THRESHOLD

According to the previous theories, the spin Meissner effect is expected to be accompanied by a specific change in spin polarization. In the presence of the external magnetic field, the minimum of the energy is achieved when Zeeman splitting is compensated by the polariton-polariton interactions. Effective screening of the external magnetic field originates from the accumulation of polaritons with spin parallel to the magnetic field, which creates an effective counteracting magnetic field. In the case of perfect equilibrium at zero temperature, the external field is completely compensated for in the ground polariton state [2]. If the full equilibrium is not achieved, we can expect a partial reduction of the energy splitting. This effect is observed below the condensation threshold (vertical shaded line in Fig. 3), when the two emission lines are still well separated but exhibit a partial reduction of the splitting. The reduction of Zeeman splitting at the pump power below the polariton laser threshold is a peculiar demonstration of the collective behavior of exciton-polaritons prior to formation of extended condensates. A similar partial reduction of the internal magnetic field is observed in a number of up-critical superconductors in the fluctuation-dominated regime [39].

At very high excitation powers the two counterpolarized signals split again, which we attribute to the loss of strong coupling and the transition to photon lasing in the dominant polarization (σ^+). The energy of the σ^+ component at this very high excitation power is fixed at the photon energy; however, the energy of the σ^- component is still increasing even though this state is not very populated. This suggests that the strong coupling is retained for excitons polarized opposite to magnetic ions [40]. The accompanying change in the emission intensity is illustrated in the Supplemental Material [31].

The variation of the degree of circular polarization (DOCP) with the excitation power at different magnetic fields is presented in Figs. 3(d)–3(f). We define DOCP as $\rho = \frac{I_{\sigma^+} - I_{\sigma^-}}{I_{\sigma^+} + I_{\sigma^-}}$, where $I_{\sigma^\pm}$ denote the emission intensities of the most populated state detected in corresponding circular polarizations. Due to the large Zeeman splitting, the emission is almost fully circularly polarized in most cases, as shown in Figs. 3(d)–3(f). However, in the vicinity of the condensation threshold and slightly below it, in the regime where the compensation of the external magnetic field occurs, the degree of circular polarization decreases. This reduction of the degree of circular polarization is due to the presence of a nonzero linear polarization splitting Ω_x and the spin Meissner effect, which reduces the effective magnetic field in the z direction [see Eq. (1)]. Above the photon lasing threshold, where the second splitting occurs, the emission builds up the spin polarization very rapidly, and the system becomes fully polarized.

We would like to comment also on the additional effect that is important for exciton-polariton condensates in semimagnetic structures, i.e., the reduction of the exciton Zeeman splitting due to the depolarization of the Mn ion subsystem by a high number of free particles created by high power and nonresonant optical excitation [41,42] (already discussed in [25]). This effect is visible at high emission angles, where we can trace out the almost pure excitonic component of the polariton state. The cross section of the polariton emission at high angles (20°) is illustrated in the Supplemental Material [31]. Figure 4(b) illustrates the Zeeman splitting of excitonlike exciton-polaritons that is observed in our structures at given excitation powers. It is reduced due to the Mn depolarization effects, but it is also clearly visible at the polariton lasing threshold (marked with gray crosses). At the highest used excitation powers the Zeeman splitting is as high as 6 meV at 6 T and is always nonzero for lower field values. Therefore, the effect of the almost complete quenching of the Zeeman splitting of the polariton condensate cannot be attributed to heating.

The obtained results are summarized by the phase diagram shown in Fig. 5. In the classical regime, at low polariton densities, the Zeeman splitting increases with the magnetic field. Complete quenching of the Zeeman splitting occurs already below the condensation threshold, and further on, in the condensed phase, it is supported to higher magnetic fields due to the spin Meissner effect. For higher pump intensities the photonic lasing occurs. This behavior is fully confirmed by our theoretical model. Note that the apparent discrepancy between theoretical and experimental photon lasing thresholds in Fig. 5 is due to the fact that in the experiment there is no clear threshold and the transition is smooth, which is clearly visible in Figs. 3(d)–3(f). We attribute this discrepancy either to the disorder in the sample (significant in CdTe-based microcavities [3,43,44]) resulting in different lasing thresholds in different parts of the sample or to the treatment of the reservoir in the model by a single equation, which does not take into account the full complexity of reservoir.

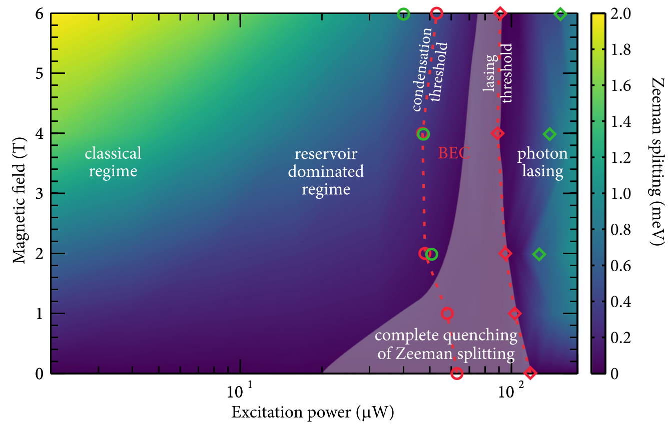

FIG. 5. Phase diagram of an exciton-polariton gas in a semimagnetic microcavity in external magnetic field. Complete suppression of Zeeman splitting is observed already below the condensation threshold in the reservoir-dominated regime. Threshold powers for condensation (photon lasing) are marked with circles (diamonds). The experimental and theoretical values are shown in red and green, respectively.

V. SUMMARY

In summary, the exciton-polariton energy splitting in magnetic field is governed by the competition of two counteracting effects: polariton-polariton interactions and the Zeeman effect. In the equilibrium spin Meissner effect these two contributions are equal, which is manifested as a reduction in the energy splitting between two states of opposite polarizations. We demonstrated that in semimagnetic microcavities, which are characterized by a giant Zeeman splitting of excitons larger than the emission linewidth, this effect is visible as a gradual reduction of the exciton-polariton Zeeman splitting even in the uncondensed state, when the system is far from thermal equilibrium. Above the condensation threshold, we observed that the energy splitting in the condensate is almost completely quenched, in accordance with the equilibrium theory predictions.

Finally, the similarity of the spin Meissner effect below the polariton lasing threshold and the partial Meissner effect in the fluctuation-dominated regime in superconductors is phenomenological: while in superconductors the effect is due to virtual Cooper pairs, in a polariton system it is governed by the spin-dependent polariton interactions with the exciton reservoir.

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