Suppression of Zeeman Splitting of the Energy Levels of Exciton-Polariton Condensates in Semiconductor Microcavities in an External Magnetic Field

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A key property of equilibrium exciton-polariton condensates in semiconductor microcavities is the suppression of the Zeeman splitting under a magnetic field. By studying magnetophotoluminescence spectra from a GaAs microcavity, we show experimentally that a similar effect occurs in a nonequilibrium polariton condensate arising from polariton parametric scattering. In this case, the quenching of Zeeman splitting is related to a phase synchronization of spin-up and spin-down polarized polariton condensates caused by a nonlinear coupling via the coherent pump state.

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The well-known Meissner effect in superconductors consists in full diamagnetic screening of the external magnetic field so that the field is exactly zero inside a type I superconductor [1]. The term "spin Meissner effect" has been introduced to describe the full paramagnetic screening effect predicted theoretically for Bose-Einstein condensates (BECs) of exciton polaritons in microcavities [2]. In the most popular GaAs- or CdTe-based planar microcavities [3–5], exciton polaritons are formed by heavy-hole excitons and have two allowed spin projections on the structure growth axis ($\pm$ 1) [6]. If no BEC is formed, the magnetic field parallel to the structure axis splits the corresponding polariton eigenstates, which represents the well-known Zeeman splitting. The spin Meissner effect consists in full suppression of the Zeeman splitting if the density of an exciton-polariton condensate exceeds a critical density dependent on the magnetic field and polariton-polariton interaction constants. In this regime, the polariton BEC is elliptically polarized. The interaction constants of polaritons having parallel spins (α_1) and antiparallel spins (α_2) are usually different, such that $\alpha_1 > \alpha_2$ [7,8]. The condensate chooses its polarization state in such a way that exactly the same energy is needed to add spin-up and spin-down polaritons due to the combined effect of spin-dependent interactions and an external magnetic field. Observation of the spin Meissner effect would require thermal equilibrium in the system, which is hard to achieve experimentally [9–11].

In this work, we observe the quenching of the Zeeman splitting with the increase of the polariton concentration in an out-of-equilibrium polariton condensate formed in the polariton optical parametric oscillator (OPO) [12–14]. While the phenomenon we observe is similar to the spin Meissner effect, its physical origin is rather different. It is associated with phase synchronization of ± 1 spin-polarized polariton condensates due to their nonlinear Josephson-type coupling via polariton-polariton spin-flip scattering processes. The synchronization leads to the coherent transfer of spin-polarized polaritons between two condensates similar to the Josephson currents in superconductors. In the stationary regime, the phase locking implies that the energies of the two condensates coincide in the case of full phase coherence in the system [15].

We studied a microcavity very similar to that of Ref. [13]. The sample was placed in a magnetic field in the Faraday geometry in a cryostat at $T \sim 12$ K. A region with Rabi splitting $2V = 6$ meV and near zero detuning between the exciton and cavity mode was investigated. The beam from a linearly polarized tunable multimode titanium sapphire laser was focused to ~ 30 μm on the sample at two different angles of incidence $\Theta_p \sim 10°$ and $\Theta_p \sim 15°$. For each excitation angle, the Zeeman splitting was studied as a function of pump power for several magnetic fields between 0 and 5 T.

At high excitation powers, resonant polariton-polariton scattering from the pump occurs in the macroscopically occupied signal state (condensate) with in-plane wave vector $k \sim 0$ and an idler state with $k \sim 2k_p$, k_p being the in-plane wave vector of the pump state [12]. For the two excitation angles the laser was tuned to energies 1.8 and $\sim$2.6 meV, respectively, above the energy of the lower polariton state at $k = 0$. These correspond to energies about 1 meV above the lower polariton state at k_p, enabling efficient excitation of parametric scattering for both σ_+ and σ_- polarized modes. The excitation conditions are illustrated in the inset in Fig. 1.

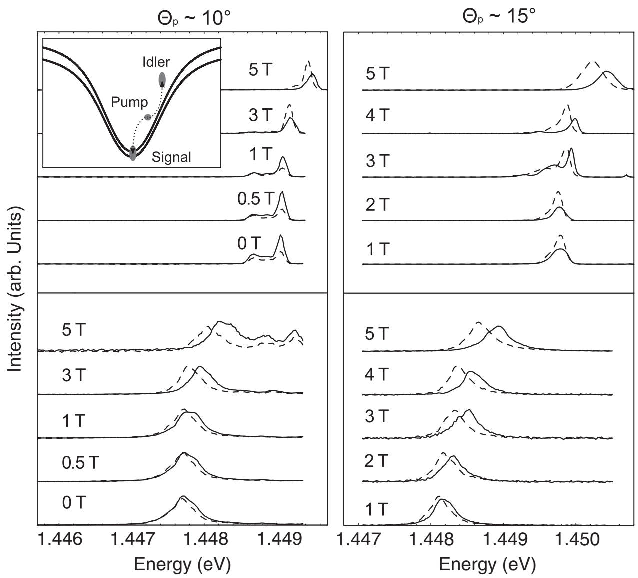

FIG. 1. Spectra of polariton emission at $k \sim 0$ recorded in two circular polarizations, σ_+ (dashed lines) and σ_- (solid lines), for various magnetic fields below (lower panels) and above (upper panels) the threshold of stimulated scattering. The inset shows a schematic diagram illustrating the polariton levels and the pump, signal, and idler states.

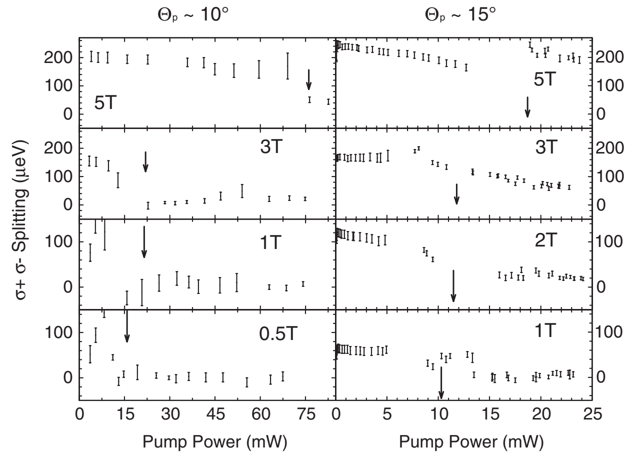

FIG. 2. Dependences of Zeeman splitting on pump power for various fields at both excitation angles. Vertical arrows indicate powers at which onset of stimulated scattering occurs.

The lower two panels of Fig. 1 show normalized spectra of polariton emission at $k \sim 0$ recorded for the two excitation angles at several magnetic fields and low excitation power below the threshold of stimulated scattering [12–14]. In this regime the main relaxation channel of polaritons from the higher to lower energy states occurs via interactions with acoustic phonons and the localized exciton reservoir. The spectra are recorded in two cross-circular polarizations σ_+ and σ_-, where the σ_+ polarization is associated with the lower energy polariton mode in the magnetic field. Despite the broad lower polariton emission (FWHM $\sim$ 0.4 meV), we are able to detect Zeeman splittings as low as 0.02 meV by using a curve-fitting procedure. The traces recorded for the σ_+ and σ_- polarizations were fit with Gaussian functions by using a commercial software package to obtain the line centers and associated uncertainties. The line center energies were subtracted to obtain the splitting.

With increasing excitation power, the intensity of both σ_+ and σ_- polarized polaritons grows superlinearly with power at all magnetic fields. This behavior arises from the onset of stimulated pair polariton-polariton scattering [12–14] from the pump, resulting in effective macroscopic occupation of the lower polariton ground σ_+ and σ_- polarized states.

The normalized spectra corresponding to the stimulated σ_+ and σ_- polarized emission at powers above threshold for $\Theta_p \sim 10°$ and $\Theta_p \sim 15°$ are shown in the upper two panels of Fig. 1. The signal emission is shifted to higher energies relative to the low power polariton emission by about 1.3–1.5 meV due to interparticle interactions between coherent OPO states and polariton interactions with the incoherent exciton reservoir [16]. The signal blueshift is proportional to the total particle density excited in the system, which is almost constant as a function of pump power above the OPO threshold [17,18]. Above the threshold, the signal lines have some multimode spectral structure, which arises from polariton spatial localization in local potential disorder [17]. We record the spectral position of the strongest individual signal mode in σ_+ and σ_- polarization as a function of pump power and magnetic field. This mode is characterized by a narrow line with FWHM $\sim$ 0.13 meV, the width of which is given by the increased coherence time of the signal condensate [17,19]. For both excitation angles there is still a finite Zeeman splitting at high magnetic fields, but the splitting is completely quenched for the lowest fields of about 1 T.

Figure 2 shows the Zeeman splitting of the signal state for several magnetic fields and both excitation angles as a function of pump power. For fields of 1 T and below, the splitting first decreases slightly with increasing pump power and then jumps suddenly to zero once the stimulated scattering threshold is reached. For $\Theta_p \sim 10°$ this behavior persists up to 3 T. At 5 T the splitting jumps from 0.20 meV down to 0.05 meV at the threshold and the splitting is only partially quenched. By contrast, for the excitation angle $\Theta_p \sim 15°$ the quenching of the Zeeman splitting is complete only for fields of 1 T and below, there being essentially no quenching at all at 5 T. There is thus a clear reduction of the Zeeman splitting in the high density system above the condensation threshold, although the degree of quenching depends on both the initial size of the Zeeman splitting and the excitation conditions.

Unlike the equilibrium system considered by Rubo, Kavokin, and Shelykh [2], where a single polariton BEC is formed, here we clearly have two nonequilibrium circularly polarized condensates. What is the mechanism of quenching of the energy splitting in this case? We argue that the system we consider is qualitatively similar to the system of two Josephson coupled polariton condensates considered theoretically by Wouters [15]. If a coherent (Josephson) coupling exists between the two condensates, their phases may by synchronized, which is achieved by a coherent transfer of polaritons from one condensate to another and vice versa. In the limit of full synchronization, the energies of the two condensates match exactly, in the stationary regime. The classical analogue of this synchronization is phase locking observed in Huygen's clocks [20]. The coherent coupling between spin-up and spin-down signal states in our case is mediated by polariton-polariton scattering via the pump state. The simultaneous spin flips of polaritons in signal and idler or signal and pump states provide a coherent transfer of signal polaritons from spin-up to spin-down states and vice versa. As we show below, this process allows a phase locking between spin-up and spin-down condensates, so that eventually a single elliptically polarized condensate may be formed. By extending earlier models [18,21] to include the polarization degree of freedom, these effects are described by coupled equations for the polarized pump ($\psi_{p,\sigma}$), signal ($\psi_{s,\sigma}$), and idler ($\psi_{i,\sigma}$) mean fields:

$$\begin{aligned} i\hbar\frac{\partial \psi_{p,\sigma}}{\partial t} = {}& E_{p,\sigma}\psi_{p,\sigma} + 2\alpha_1|X_p|^2\psi^*_{p,\sigma}\psi_{s,\sigma}\psi_{i,\sigma} \\ & + fe^{i\omega t} + \alpha_1|X_p|^2(|\psi_{p,\sigma}|^2 + 2|\psi_{s,\sigma}|^2 \\ & + 2|\psi_{i,\sigma}|^2)\psi_{p,\sigma} + \alpha_2|X_p|^2[(|\psi_{p,-\sigma}|^2 \\ & + |\psi_{s,-\sigma}|^2 + |\psi_{i,-\sigma}|^2)\psi_{p,\sigma} \\ & + (\psi^*_{s,-\sigma}\psi_{s,\sigma} + \psi^*_{i,-\sigma}\psi_{i,\sigma})\psi_{p,-\sigma} \\ & + (\psi_{s,\sigma}\psi_{i,-\sigma} + \psi_{s,-\sigma}\psi_{i,\sigma})\psi^*_{p,-\sigma}], \end{aligned} \tag{1}$$

$$\begin{aligned} i\hbar\frac{\partial \psi_{s,\sigma}}{\partial t} = {}& E_{s,\sigma}\psi_{s,\sigma} + \alpha_1|X_s|^2\psi^*_{i,\sigma}\psi^2_{p,\sigma} \\ & + \alpha_1|X_s|^2(|\psi_{s,\sigma}|^2 + 2|\psi_{p,\sigma}|^2 + 2|\psi_{i,\sigma}|^2)\psi_{s,\sigma} \\ & + \alpha_2|X_s|^2[(|\psi_{p,-\sigma}|^2 + |\psi_{s,-\sigma}|^2 + |\psi_{i,-\sigma}|^2)\psi_{s,\sigma} \\ & + (\psi^*_{p,-\sigma}\psi_{p,\sigma} + \psi^*_{i,-\sigma}\psi_{i,\sigma})\psi_{s,-\sigma} \\ & + \psi_{p,\sigma}\psi_{p,-\sigma}\psi^*_{i,-\sigma}], \end{aligned} \tag{2}$$

$$\begin{aligned} i\hbar\frac{\partial \psi_{i,\sigma}}{\partial t} = {}& E_{i,\sigma}\psi_{i,\sigma} + \alpha_1|X_i|^2\psi^*_{s,\sigma}\psi^2_{p,\sigma} \\ & + \alpha_1|X_i|^2(|\psi_{i,\sigma}|^2 + 2|\psi_{p,\sigma}|^2 + 2|\psi_{s,\sigma}|^2)\psi_{i,\sigma} \\ & + \alpha_2|X_i|^2[(|\psi_{p,-\sigma}|^2 + |\psi_{s,-\sigma}|^2 + |\psi_{i,-\sigma}|^2)\psi_{i,\sigma} \\ & + (\psi^*_{p,-\sigma}\psi_{p,\sigma} + \psi^*_{s,-\sigma}\psi_{s,\sigma})\psi_{i,-\sigma} \\ & + \psi_{p,\sigma}\psi_{p,-\sigma}\psi^*_{s,-\sigma}]. \end{aligned} \tag{3}$$

The bare energies $E_{n,\sigma}$ are given by the standard mode coupling equation [22], where the exciton energies are modified by the bare Zeeman splitting. Both exciton and cavity photon energies should also have a negative imaginary part, representing particle decay. f represents a continuous wave, linearly polarized, coherent pump with angular frequency ω. X_m are the excitonic Hopfield coefficients [22]. With parameters relevant for our experiment, Eqs. (1)–(3) can be solved numerically by starting from the initial conditions that the amplitude and phase of the six states in the model are random and the intensity of the states is very small.

For parameters corresponding to our experiment, Fig. 3(a) shows the dependence of the relative phase between σ^+ and σ^- polarized signal components after the pump is switched on for different initial conditions (different random phases). Remarkably, on a time scale on the order of a few hundred picoseconds, the σ^+ and σ^- phases lock to a fixed phase difference, independent of the initial condition. The locking of the phases requires the σ^+ and σ^- polarized signal components to have the same energy, such that the Zeeman splitting is, by definition, suppressed.

In the case of no interactions between oppositely polarized spins, $\alpha_2 = 0$, the phase locking does not take place. This is shown in Fig. 3(b), where the phase difference between σ^+ and σ^- polarized signal components is random, depending on the initial condition, and is not constant in time. Since there is no phase locking, one can expect a Zeeman splitting to remain in the system, which is reasonable given that σ^+ and σ^- polarizations are independent.

For nonzero α_2, the phase-locking mechanism has little sensitivity to the pump power, in agreement with the experimental results for $\Theta_p \sim 10°$, where suppression of Zeeman

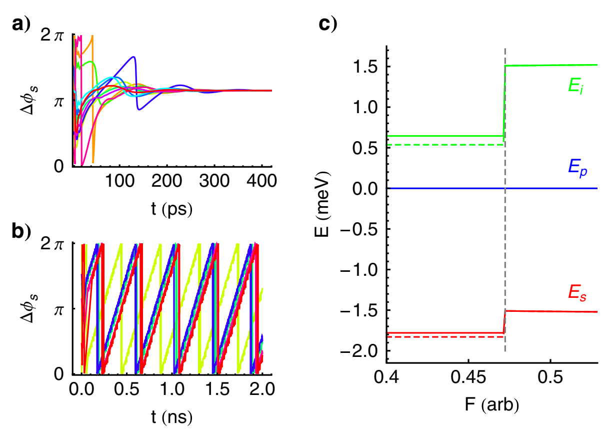

FIG. 3 (color online). (a) Time dependence of the relative phase of σ^+ and σ^- polarized signal components for different initial conditions and $\alpha_2 = -0.05\alpha_1$. (b) The same as in (a) for $\alpha_2 = 0$ showing that phase locking does not take place. (c) Dependence of the pump, signal, and idler energies on the pump intensity for σ_+ (dashed lines) and σ_- (solid lines) polarizations with $\alpha_2 = -0.05\alpha_1$. The vertical line indicates the parametric scattering threshold. The dispersion was taken with a Rabi splitting of 6 meV and a cavity photon effective mass $\sim 5 \times 10^{-5}$ of the free electron mass. The photon and exciton linewidths were taken as 0.1 and 0.5 meV, respectively. The pump was taken with an angle of incidence of $\Theta_p = 10°$ and an energy 1 meV above the bare lower polariton energy at the same angle. $B = 1$ T.

splitting was observed for a range of pump powers. A typical power dependence of the energies is shown in Fig. 3(c). The signal blueshift in the theory above the parametric threshold is about 0.5 meV smaller than that observed in the experiment. This disagreement can be attributed to inhomogeneous broadening of the exciton leading to higher losses in the idler and population of the incoherent reservoir. In addition, a stronger blueshift may arise from a reduction of the polariton Rabi splitting due to partial screening of the exciton resonance at high pump density. These mechanisms were not taken into account in our model.

In a qualitative picture, the energy of coupling between coherent cross-polarized polariton modes is proportional to $\alpha_2 N_{\rm coh}$, where $N_{\rm coh}$ is the total coherent polariton density in the OPO states, which is given mainly by the pump population. Taking into account that the signal blueshift above threshold is of the order of $\alpha_1 N_{\rm coh} \sim 1$–$1.5$ meV, we deduce that $\alpha_2 N_{\rm coh} \sim 0.1$ meV. The efficient mode synchronization leading to quenching of Zeeman splitting above the threshold occurs in the case when $\alpha_2 N_{\rm coh}$ is larger than the Zeeman splitting below the threshold, which is consistent with experimental observations in Figs. 1 and 2. At the larger excitation angle (15°) studied in this work, the pump energy is tuned to an energy closer to the exciton level by about 1 meV than at the smaller pump angle (10°). In this situation the pump state can be strongly depleted due to absorption by an incoherent excitonic reservoir, which consists of localized or dark excitons and has an exponentially increasing density of states with increasing energy towards the exciton level [16]. As a result of such depletion, the coupling strength between the σ_+ and σ_- polarized signal states is decreased at $\Theta_p \sim 15°$, leading to quenching of Zeeman splitting at smaller B fields of 1–2 T in this case.

In conclusion, by studying the magnetophotoluminescence spectra of a semiconductor microcavity in the OPO configuration, we observed experimentally the density-dependent renormalization of the Zeeman splitting, which is in agreement with calculations based on the mean-field coupled mode equations. We find that the Zeeman splitting is suppressed above the threshold for parametric oscillation. This is associated with a phase synchronization between spin-up and spin-down polariton condensates caused by coherent spin-flip processes mediated by polariton parametric scattering. This effect has the same origin as the recently proposed polariton spin Josephson effect [23].

We note that, following the submission of our manuscript, the suppression of Zeeman splitting was also reported for incoherently pumped polariton condensates in semiconductor microcavities [24].

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