Magnon-magnon interactions in a room-temperature magnonic Bose-Einstein condensate

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(Received 12 April 2017; revised manuscript received 17 July 2017; published 30 August 2017)

The Bose-Einstein condensate of magnons (mBEC) that is formed at room temperature in parametrically pumped magnetic films is doubly degenerate: it is formed simultaneously in two spectral minima corresponding to the lowest-energy magnons propagating in opposite directions along the in-plane bias magnetic field. In this work the interactions of magnons in the mBEC are studied both theoretically and experimentally. It is shown by direct calculation that the magnons residing in each of the degenerate spectral minima of mBEC form a practically ideal magnon gas, as the attractive self-interaction between these magnons is very weak. At the same time, the interaction between the magnons residing in different spectral minima, corresponding to opposite directions of the magnon wave vector, is relatively strong and repulsive, leading to a repulsive total intermagnon interaction. By measuring the spectral characteristics of the mBEC it is shown that with increased magnon density the energy per magnon in the mBECs increases, thus confirming experimentally that the net intermagnon interaction in a doubly degenerate mBEC is repulsive.

DOI: 10.1103/PhysRevB.96.064438

I. INTRODUCTION

A Bose-Einstein condensate (BEC) is a system of (quasi)particles in which most of the particles occupy a single quantum state in the momentum space [1,2]. During the last 20 years BECs were observed in many kinds of gases, consisting of different real atoms, as well as of quasiparticles, namely, polaritons [3,4], excitons [5], and magnons. In particular, the BEC of magnons (mBEC) was observed in a ^{3}He superfluid at extremely low temperatures [6], quantum magnets [7], and ferrite films at room temperature [8–11]. It was found that microwave magnons created by external parametric pumping after thermalization form a state of coherent precession with a phase and frequency independent of the external pumping signal.

In the process of a BEC the density of quasiparticles drastically increases [11], and the interactions between the quasiparticles start to play an important role in the dynamics of the BEC. Usually, the interaction between atoms in the BEC is described by a "hard-sphere" model, introducing a nonlinear energy term in the Hamiltonian in the form [12–14]

$$W_{\text{int}}(\mathbf{r}' - \mathbf{r}) = \frac{4\pi\hbar^2 a}{m}\delta(\mathbf{r}' - \mathbf{r}) = g\delta(\mathbf{r}' - \mathbf{r}), \qquad (1)$$

where m is the particle mass and a is the scattering length [14]. The interparticle interaction coefficient g [12] (called in wave physics the nonlinear coefficient) is directly related to the scattering length a. This coefficient, g, defines both the strength and the character of the interparticle interaction. If $g < 0$, the interaction is attractive (while if $g > 0$, it is repulsive), meaning that the addition of particles at the same volume decreases (increases) the energy per particle in the BEC. For the BEC of atoms the change in sign of the scattering length a (and coefficient g) from positive to negative results in a global collapse of an untapped condensate [15,16]. In fact, the discovery of the Feshbach resonance [17] has recently opened the way to study quantum liquids and coherent states of matter in widely varying circumstances with positive, negative, and zero values of the scattering length a (and, correspondingly, of g) in a single experiment [18,19]. The potential (1) can still be used for modeling the interaction between the quasiparticles, but in the case of quasiparticles the mass m denotes an effective mass of a quasiparticle. The effective quasiparticle mass can be negative (e.g., in periodic potentials [4]), but regardless of the sign of the effective mass, the sign of the interaction coefficient g defines whether the interaction is attractive or repulsive and therefore serves as a good indicator of the possibility of the BEC collapse.

At the same time, it is well known that the interaction between magnons in relatively thick ferromagnetic films is attractive [20]. Thus, the energy per particle of the condensate should decrease with increasing density, which, in principle, should lead to a collapse of the mBEC [12]. However, in experiments [8–11], the observed mBEC was stable. A possible explanation for this contradiction can be found by taking into account an important peculiarity of the mBEC: the magnonic spectrum in an in-plane magnetized film has two degenerate minima with wave numbers k_{BEC} and $-k_{\text{BEC}}$ (see Fig. 1). Under a parametric excitation [8–11,20] magnons have an equal probability to populate both minima, forming *two* mBECs simultaneously. The nonlinear dynamics in this mixture of two mBECs is determined not only by the magnon self-action (which is attractive) but also by the mutual interaction (cross interaction) of magnons in both minima.

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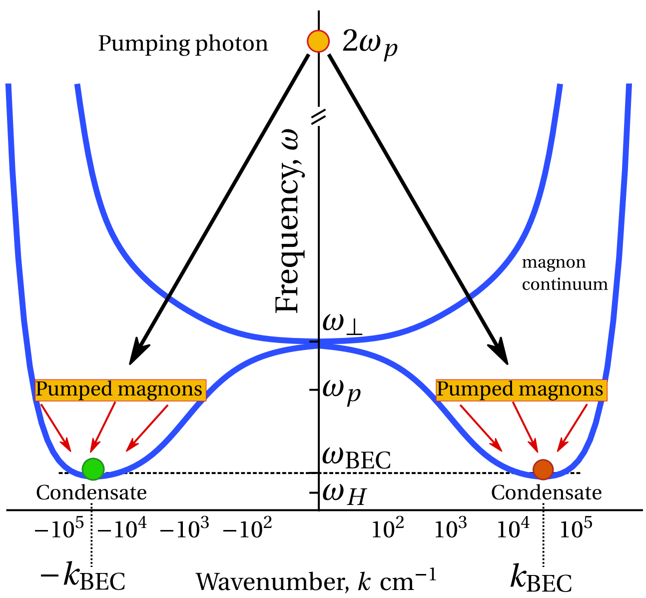

FIG. 1. Schematic representation of the magnon dispersion $\omega_0(k)$ in an in-plane magnetized magnetic film for magnons propagating along the bias magnetic field (backward volume magnetostatic wave geometry), illustrating formation of a two-component BEC of magnons in the two degenerate spectral minima. Parametric pumping at the frequency $2\omega_p$ excites magnons of the frequency ω_p in the magnon spectrum (orange shaded areas), which, due to the thermalization in a magnon gas, form a two-component BEC at the spectral minima with wave numbers $\pm k_{BEC}$ and the same frequency ω_{BEC}, indicated by green and red dots. Here $\omega_H = \gamma\mu_0 H$, $\omega_\perp = \sqrt{\omega_H(\omega_H+\omega_M)}$, $\omega_M = \gamma\mu_0 M_s$, γ is the gyromagnetic ratio, H is the applied magnetic field, and M_s is the saturation magnetization.

Indeed, if this cross interaction is repulsive and strong, then, under certain conditions, it can become more important than the attractive self-action, leading to the resultant repulsion between the interacting magnons and increase of the energy per particle in both condensates [21,22]. Fortunately, in the case of a mBEC by measuring the mBEC frequency with the increasing mBEC density it is possible to directly access the energy of magnons in the mBEC [23] and check this hypothesis.

Calculations show, however, that this nonlinear frequency shift is quite low ($\delta\omega/(2\pi) \approx 3$ MHz) at the mBEC frequency of $\omega/(2\pi) \approx 6$ GHz. To check experimentally the above-presented hypothesis of the dominant role of the cross interaction between magnons with opposite wave vectors in the nonlinear frequency shift of the mBEC we built a setup based on the recently developed heterodyne technique which employs the magneto-optical Kerr effect [24]. This technique has a frequency resolution of $\delta\omega/\omega = 10^{-8}$, which is sufficient to reliably detect the expected frequency shift of the mBEC. Using this setup, we measured the frequency of the mBEC while varying the pumping power and found that the frequnecy of the mBEC linearly increases with the pumping power (proportional to the magnon density), confirming the repulsive nature of the resultant intermagnon interaction in the mBEC.

II. THEORY OF MAGNON INTERACTIONS IN THE mBEC

We consider a mBEC residing in two spectral minima and having the wave vectors k_{BEC} and $-k_{BEC}$. This system can be considered a gas made of particles of two different kinds (species) [12,25]. The dynamics of such a two-component BEC can be understood using the same model of hard spheres but assuming that the interaction coefficients are different for the particles of different kinds: $W_{\rm int}^{i,j}(\mathbf{r}-\mathbf{r}') = g^{i,j}\delta(\mathbf{r}-\mathbf{r}')$, where $i,j = \{1,2\}$. In particular, the case of an untrapped BEC formed from a two-component gas of interacting particles (or quasiparticles) can be described by equations of coupled harmonic oscillators, which in the representation of second quantization have the form [11]

$$i\hbar\dot{b}_1 = \left(\hbar\omega_0 + g\frac{b_1^* b_1}{V} + 2g^{12}\frac{b_2^* b_2}{V}\right)b_1,$$
$$i\hbar\dot{b}_2 = \left(\hbar\omega_0 + g\frac{b_2^* b_2}{V} + 2g^{12}\frac{b_1^* b_1}{V}\right)b_2, \tag{2}$$

where b_i is the complex amplitude of (quasi)particles of kind i, normalized by the condition $b_i^* b_i = N_i$, N_i is the number of particles of the ith type, ω_0 is the lowest frequency of the single-particle spectrum, and V is the volume occupied by the two-component BEC.

The presence of an interaction between the particles in the BEC results in the change in the net energy per particle in the system, which can be measured experimentally. Assuming that the two-component BEC is spatially uniform with the density $\rho_i = b_i^* b_i/V = \rho$, it is possible to evaluate the nonlinear frequency shift of the two-component BEC as

$$\delta\omega = \omega - \omega_0 = (g/\hbar)\rho + (2g^{12}/\hbar)\rho. \tag{3}$$

Thus, if $g^{12} > 0$ and $g^{12} \gg |g|$, the net energy per particle $E = \hbar\omega$ increases with the increase in the BEC density, and the collapse of the BEC is avoided.

Magnons in a mBEC interact with each other in the nonlinear processes that conserve both energy and linear momentum [20]. Thus, we are interested in only the processes between the magnons either having the same wave vector $k,k \to k,k$ [$g(k)$ is the self-interaction of magnons in one spectral minimum] or having opposite wave vectors $k,-k \to k,-k$ [$g^{12}(k)$ is the cross interaction of magnons belonging to different spectral minima]. To calculate the magnon-magnon interaction coefficients $g(k)$ and $g^{12}(k)$, we used a standard Hamiltonian approach [20–22,25–27] (see the Supplemental Material [28] for details). For relatively large magnon wave numbers k corresponding to the spectral minima, where the mBEC is formed [note that these wave numbers are still substantially smaller than the reciprocal vectors of the yttrium iron garnet (YIG) crystal lattice], the magnetization precession has a practically circular polarization. In this approximation, it is possible to obtain the following approximate expressions for the above coefficients, describing magnons in the in-plane magnetized magnetic film, in a simple analytical form:

$$g(k) \approx -\frac{\omega_0(k)-\omega_H}{\omega_M}\mu_0\hbar^2\gamma^2,$$
$$2g^{12}(k) \approx \left[4\lambda_{\rm ex}^2 k^2 + Q(2k)\right]\mu_0\hbar^2\gamma^2 + 2g(k), \tag{4}$$

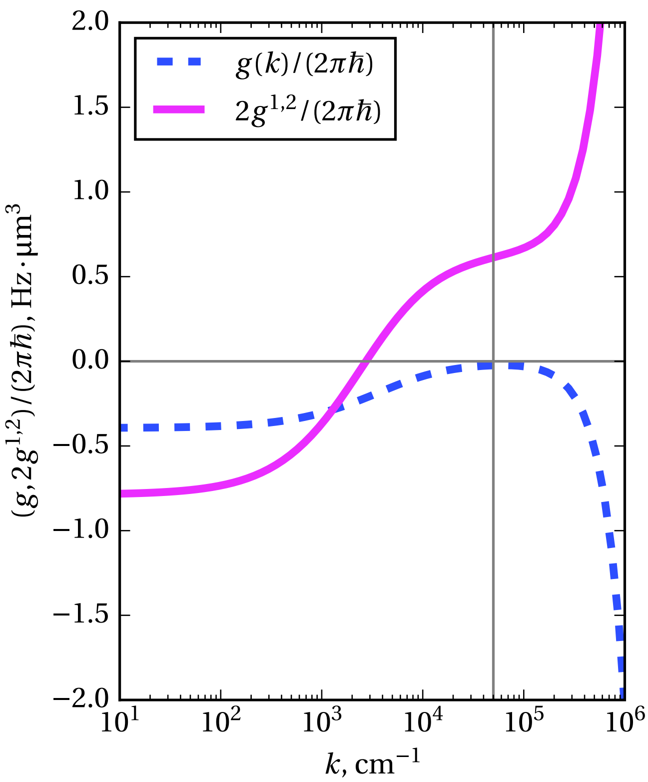

FIG. 2. Magnon interaction coefficients versus wave numbers $g(k)$, describing the self-action of magnons inside one of the spectral minima (dashed blue curve), and $g^{12}(k)$, describing the interaction of magnons with opposite wave vectors and belonging to different spectral minima (see Fig. 1). The nonlinear coefficients are presented in units of frequency.

where $\omega_0(k)$ is the dispersion of magnons in the fundamental (uniform along the film thickness) mode, $\omega_M = \gamma\mu_0 M_s$, M_s is the saturation magnetization of the ferromagnetic film, $\omega_H = \gamma\mu_0 H$, H is the applied magnetic field, γ is the gyromagnetic ratio, λ_{ex} is the exchange length, $Q(k) = (-1 + kL + e^{-kL})/(kL)$ is the matrix element of the dipole-dipole interaction, and L is the film thickness. More accurate full expressions for these coefficients that take into account the real elliptical polarization of the magnetization precession are provided in the Supplemental Material [28], and the wave-number dependences of these more accurately calculated coefficients are presented in Fig. 2.

It is clear from Eq. (4) and Fig. 2 that the self-action coefficient $g(k)$ is negative in the full interval of the magnon wave-number variation since $\omega_0(k) > \omega_H$ (see Fig. 1), so the interaction between the magnons belonging to the same spectral minimum is always attractive. At the point of the mBEC formation, this attractive interaction is, however, *very weak*, making the single-component magnon gas practically *ideal*.

In contrast, the coefficient of interaction $g^{12}(k)$ between the magnons belonging to different spectral minima behaves in a different way: it changes sign from negative to positive with the increase in the magnon wave number. In particular, at the point $k = k_{BEC}$, where the mBEC is formed, the coefficient $g^{12}(k)$ is positive and is much larger than $g(k)$. Thus, the strong repulsive interaction between the magnons belonging to different spectral minima with the wave numbers k_{BEC} and $-k_{BEC}$ is dominant in this two-component mBEC and determines the overall nonlinear behavior of the mBEC. It is also worth noting that the magnitude of the self-action coefficient $g(k)$ rapidly increases when the magnon wave number is shifted away from the spectral minima. This increased intermagnon interaction is the origin of the strong four-magnon scattering leading to a fast thermalization of magnons, which is necessary for the formation of a mBEC in a gas of parametrically pumped magnons in the first place [8,9,11].

For a typical density of a mBEC of $\rho = 5 \times 10^6 \mu\text{m}^{-3}$ [11,29] the frequency shift associated with the magnon self-action is $g(k_{BEC})\rho/(2\pi) \approx -0.14$ MHz, which is comparable to the intrinsic linewidth of magnons in a ferrite (YIG) film. At the same time, the coefficient $g^{12}(k)$, describing the cross interaction between the different components of the mBEC, is positive and has a significant magnitude, thus totally dominating the nonlinear dynamics of magnons in a two-component BEC of magnons. The frequency shift associated with this cross interaction is $2g^{12}(k)\rho/(2\pi) \approx 3.0$ MHz.

Thus, by a direct measurement of the nonlinear frequency shift of the mBEC one can check the above-presented hypothesis and find out experimentally whether the cross interaction of magnons belonging to the two spectral minima in the mBEC is, indeed, repulsive and dominant. The absolute value of the measured nonlinear frequency shift is proportional to the magnon density (3), which cannot be measured directly and independently with sufficient accuracy. However, (i) the magnon density was estimated previously by measuring the chemical potential in the mBEC [29], and (ii) the interaction coefficients describing the self- and cross interactions between the magnons existing in condensates with positive k_{BEC} and negative $-k_{BEC}$ wave vectors have different signs. Thus, their effect on the total experimentally measured nonlinear frequency shift can be elucidated from the qualitative behavior of this shift (positive or negative).

III. EXPERIMENTAL MEASUREMENT OF THE NONLINEAR FREQUENCY SHIFT IN THE mBEC

The predicted magnitudes of the nonlinear frequency shifts of a mBEC above are rather small: they constitute only a small fraction ($\delta\omega/\omega \approx 10^{-4}$) of the mBEC frequency ω corresponding to the degenerate magnon spectral minima. The measurement of such a small frequency shift for magnons having a rather large wave number constitutes a very challenging experimental problem since the traditional high-frequency-resolution microwave spectroscopy is not sensitive to magnons with large wave numbers. The state-of-the-art Brillouin light scattering (BLS) setups, while having very high amplitude sensitivity, usually have a frequency resolution of upward of 50 MHz [7,29], which is definitely not sufficient for studies of such delicate nonlinear effects. To meet this experimental challenge, we used a recently developed heterodyne magneto-optical Kerr-effect (MOKE) frequency detection technique [23].

The schematic of the experimental setup (described in detail in Ref. [23]) is shown in Fig. 3(a). A microstrip-based

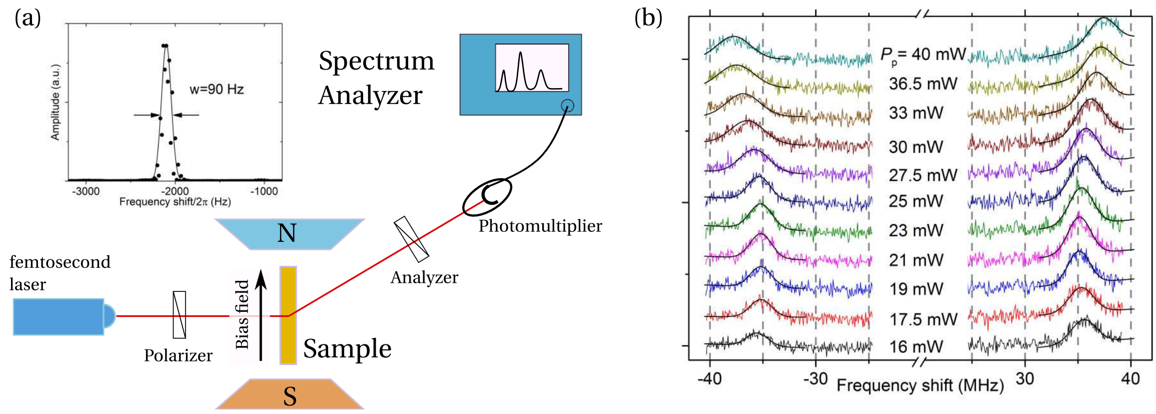

FIG. 3. (a) Experimental setup. A beam from a femtosecond laser is focused on a ferrite film magnetized by an electomagnet. The scattered light is collected by a photomultiplier, and the output signal is analyzed by a spectrum analyzer. The inset shows a measured reference spectrum with a linewidth of 90 Hz. (b) Measured spectra of the magnonic BEC for different values of the pumping power. Solid black lines indicate fitting done to reveal the frequency.

microwave resonator parametrically pumps magnons in an YIG film. The sample is placed in a static magnetic field H. Linearly polarized laser pulses with a wavelength of 490 nm (after frequency doubling), a duration of 100 fs, and a highly stabilized repetition frequency $\omega_L/(2\pi) = 82.379$ MHz are focused onto the sample near the pumping resonator. The Bose-Einstein condensation of magnons in ferromagnetic films takes place for magnons having wave numbers $k_{\rm BEC} \approx 5 \times 10^4\,{\rm cm}^{-1}$. This range of magnon wave numbers is fully accessible for the experimental technique of Brillouin light scattering with a light wavelength of 490 nm [10,23,29]. After passing through the sample, the light is sent through the second polarizer (analyzer) and directed to a photomultiplier with a frequency band of about 100 MHz. The output electronic signal is analyzed by a radio-frequency spectrum analyzer. The YIG film used in our experiments was fabricated by liquid-phase epitaxy on a gadolinium gallium garnet (GGG) substrate. Both materials (YIG and GGG) are optically transparent, which allowed us to analyze the translucent light beam. The collection optic elements are mounted on a rotational arm with the rotation axis at the condensate position, which enables detection and analysis of light scattered by the magnons with wave number $k_{\rm BEC} = 5 \times 10^4\,{\rm cm}^{-1}$. Due to the MOKE, the output light becomes modulated in amplitude with the frequency of the detected magnons $\omega(k)$, which is superimposed with the repetition frequency ω_L. The Fourier spectrum of the output signal consists of equidistant harmonics $n\omega_L$, each of them having two (Stokes and anti-Stokes) satellites with frequency shifts $n\omega_L \pm \Delta\omega$, where $\Delta\omega = |\omega(k) - N\omega_L|$, with N being the nearest integer to $\omega(k)/\omega_L$. Thus, a standard heterodyne scheme is used to reach a resolution of $\delta\omega/(2\pi) \approx 100$ Hz, as demonstrated by independent experiments [23] [see the inset in Fig. 3(a)].

To obtain information about the nonlinear frequency shift of the BEC, several spectra were recorded at a constant magnetic field of $H_0 = 113.91$ kA/m but for different values of the pumping power P_p [see Fig. 3(b)]. It is clear from Fig. 3(b) that the central frequency of the signal obtained from the magnons situated in the spectral minimum at first drops with increasing power, but at a certain threshold power of $P_{\rm th} \approx 19$ mW it starts to increase, as demonstrated in Fig. 4(a) in detail. The initial drop in the magnon frequency is connected to the fact that before the condensation of magnons starts to happen, the chemical potential of the magnon gas increases, and the particle-energy distribution in a gas of magnons changes, resulting in a continuously increasing population of states close to the spectral minima [8].

Then, for $P > P_{\rm th}$, the mBEC is formed, so the magnons become "densely packed" near the spectral minima, and the interactions between them start to strongly influence the magnon frequency and play a significant role in the magnon dynamics. Although the exact quantitative relation between the pumping power and the density of the mBEC strongly depends on the experimental details, it has been shown in previous experiments performed at similar moderate magnitudes of the pumping power that the BEC density is approximately proportional to $P_p - P_{\rm th}$ if $P_p > P_{\rm th}$ [21]. It is seen from Fig. 4(a) that the measured magnon frequency shift for $P_p > P_{\rm th}$ increases linearly with the pumping power, therefore confirming the theoretical dependence (3) and (4). On the other hand, the magnon-magnon interaction should also increase the linewidth of the fundamental magnonic mode [30]. The measured dependence of the magnon linewidth on the pumping power is shown in Fig. 4(b). Indeed, before the threshold power $P_{\rm th}$ of the BEC formation, the linewidth of the magnon mode drops with the increase in power, but above the mBEC threshold it starts to increase due to the mBEC nonlinearity. It is important to note that the qualitative change in slope in the power dependences of both the magnon nonlinear frequency shift and the magnon linewidth takes place *at the same magnitude* of the pumping power, leaving a "smoking gun" of both the formation of a mBEC at this power and the appearance of a strong repulsive interaction between the magnons residing in the two degenerate spectral minima.

IV. DISCUSSION AND CONCLUSIONS

The presented theory and experiment clearly demonstrate that the per particle energy of a mBEC increases with the

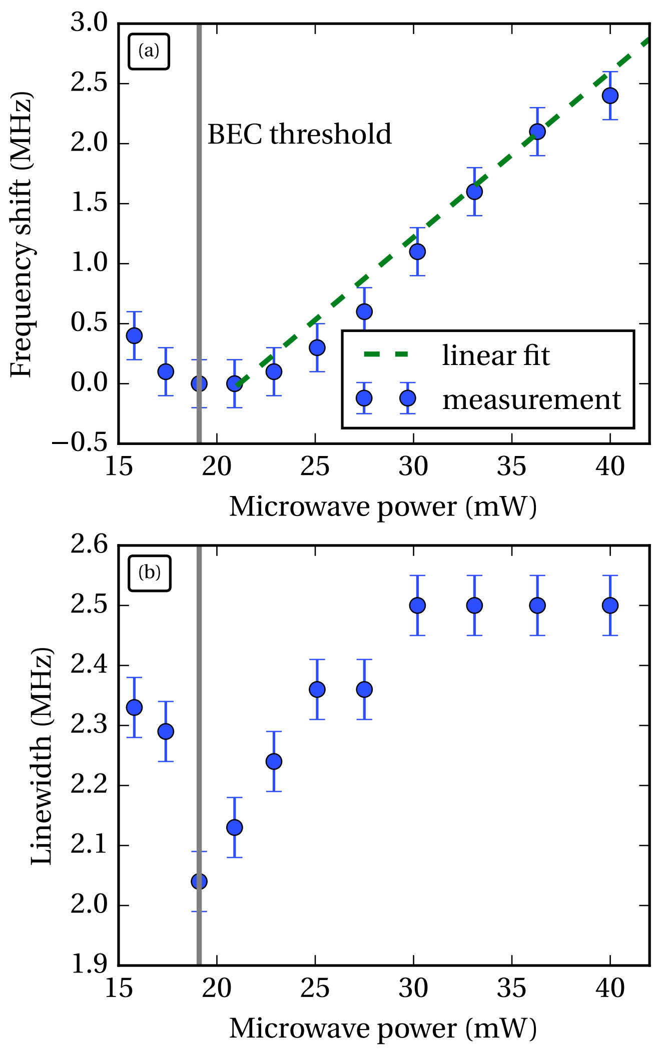

FIG. 4. Measured (a) frequency shift and (b) linewidth of the magnonic BEC as a function the pumping power (dots). The gray line corresponds to the threshold power of the BEC formation, and the green dashed line is a linear trend for the frequency dependence on the pumping power in the supercritical regime $P_p > P_{\text{th}}$.

magnon density. Thus, the mBEC cannot collapse explosively with a rapid increase in the magnon density. However, other types of instabilities can take place in the mBEC.

In this paper we considered magnon-magnon interactions of spatially uniform mBECs. The underlying physics and, in particular, the mBEC stability analysis become much more complicated if a possible spatial nonuniformity of the magnon wave function in the mBEC is taken into account. In particular, the repulsive cross interaction between the magnons residing in the degenerate spectral minima can lead to phase separation in a multicomponent BEC [31,32]. Such a phase separation can lead to the formation of "domains" of magnons with different densities of magnons having opposite wave numbers and, as a consequence, to the reduction of the per-particle energy. A strong phase separation of magnons in the mBEC is not observed in our experiments with spatial resolution [22,33], possibly because the relaxation time for magnons (0.5 μs for YIG) is of the same order as the phase-separation time ($2\pi/\delta\omega = 0.3$ μs), and the pronounced domains cannot be formed under the relatively low pumping rate used in our experiments.

In general, detailed information about the properties of the mBEC at room temperature can be found in the review in Ref. [34].

In conclusion, we studied both theoretically and experimentally the interaction of magnons in a magnonic condensate. We were able to show that the repulsive interaction between the magnons with opposite wave vectors residing in two degenerate spectral minima of a mBEC is dominant in comparison to the weak attractive self-action of magnons residing in each of the spectral minima.

Using a measurement setup that combines the magneto-optical Kerr-effect with a homodyne detection technique and provides a record-high frequency resolution, we experimentally measured the nonlinear frequency shift and the linewidth of the mBEC at room temperature and confirmed experimentally that the energy of the mBEC increases with the increase of the pumping power, creating the microwave magnons in the studied system.

ACKNOWLEDGMENTS

This work was supported in part by the Deutsche Forschungsgemeinschaft, the program Megagrant 14.Z50.31.0025 of the Russian Ministry of Education and Science, Russian Science Foundation under Grant No. 14-19-00760, the National Science Foundation of the United States (Grants No. EFMA-1641989, No. ECCS-1708982), a grant from DARPA (Grant No. M3IC # W911NF-17-C-0031), and a grant from the Center for NanoFerroic Devices (CNFD) and Nanoelectronics Research Initiative (NRI). This work was also supported by the Foundation for Fundamental Research on Matter (FOM), de Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO), European Unions Seventh Framework Programme (FP7/2007-2013) Grant No. 281043 (FemtoSpin), and European Research Council Grant No. 339813 (Exchange).

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