

Correlation-Enhanced Interaction of a Bose-Einstein Condensate with Parametric Magnon Pairs and Virtual Magnons

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Nonlinear interactions are crucial in science and engineering. Here, we investigate wave interactions in a highly nonlinear magnetic system driven by parametric pumping leading to Bose-Einstein condensation of spin-wave quanta—magnons. Using Brillouin light scattering spectroscopy in yttrium-iron garnet films, we found and identified a set of nonlinear processes resulting in off-resonant spin-wave excitations—virtual magnons. In particular, we discovered a dynamically strong, correlation-enhanced four-wave interaction process of the magnon condensate with pairs of parametric magnons having opposite wave vectors and fully correlated phases.

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Nonlinear wave interactions govern the behavior of various systems in nature, including processes in the Earth's ocean and atmosphere [1], in stars [2], and even the evolution of the Universe [3]. In the field of magnetism, the use of nonlinear phenomena opens promising prospects for applications of magnonics and spintronics in neuromorphic computing, microwave data processing, and nanoscale wave logic circuits [4–11]. Spin waves, or magnons, in magnetically ordered materials are highly nonlinear compared with, for example, phonons or photons in solids. One of the best systems for studies of nonlinear spin-wave phenomena is the single-crystal ferrimagnetic yttrium iron garnet (YIG, $\text{Y}_3\text{Fe}_5\text{O}_{12}$) material [9,12,13]. A strong magnon nonlinearity, combined with a high quality factor of magnons in YIG, facilitates the creation, registration, and study of various interaction processes.

The phenomenon of Bose-Einstein condensation of magnons at the bottom of their frequency spectrum, observed in YIG films [14–20], also develops through the nonlinear scattering of magnons [21]. In our experiments, this condensation was achieved by the parametric pumping [22] of magnons with microwave radiation [14,15,20,21,23–25].

Here, we reveal several nonlinear processes that involve not only “real” quasiparticles—the eigenmodes of the

medium—but produce, as a final result, virtual quasiparticles—out-of-resonance waves—caused by various types of nonlinear interactions. The most nontrivial process involves a pair of parametrically excited magnons and a Bose-Einstein condensate (BEC). This process is enhanced by full-phase correlations in parametric magnon pairs with opposite wave vectors $\mathbf{q}$ and $-\mathbf{q}$.

Previously, virtual quasiparticles (e.g., virtual magnons) appeared in the theory of nonlinear waves [26–29] only as mathematical abstractions, mediating interactions between “real” quasiparticles. At the same time, the experimental observation of virtual quasiparticles is crucial for the identification of possible physical processes in nonlinear wave systems. This is especially critical when, for various reasons, the experimental methods do not allow one to observe the real quasiparticles involved in these processes.

In our studies, we used Brillouin light scattering (BLS) spectroscopy [30,31] employing a triple-pass tandem Fabry-Pérot interferometer [32]. Its high sensitivity enables the detection of a tiny number of magnons even at thermal noise level, and the frequency resolution (as low as 50 MHz) is quite sufficient for experiments with magnons in the 3–15 GHz range, which was explored in our investigation. The 532 nm laser beam is focused into a 20 μm diameter probing spot in the parametrically pumped area of the YIG film. By setting the incidence angle $\Theta_{q\parallel}$ of this beam [33], the wave vectors of magnons propagating in the YIG film plane along the line of its intersection with the beam incidence plane are selected. Since, in our case, the incident plane is oriented along the direction of the bias magnetic field $\mathbf{H}$, magnons propagating in the same direction with wave vectors $\mathbf{q}_{\parallel}\parallel\mathbf{H}$ are detected. The change

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in the incidence angle from 0 to the maximal value of 58° in our experiment corresponds to the change in the detectable wave vectors from 0 to $2 \times 10^5 \text{ cm}^{-1}$. The wave vector resolution in our experiments is of the order of $\pm 10^3 \text{ cm}^{-1}$ [34].

Microwave pumping at 13.2 GHz is applied to the in-plane magnetized 6.7 μm -thick YIG film using a microstrip resonator of 50 μm width. In order to achieve high magnon densities $n(\omega, \mathbf{q})$, the pumping is supplied in pulses of up to 20 W and duration of 1 μs . A sufficiently low repetition rate of 1 kHz maintains a thermal quasiequilibrium from pulse to pulse.

Our experimental results for the BLS intensity spectra $I(\omega, q_{\parallel})$, proportional to $n(\omega, q_{\parallel})$, are shown in Fig. 1. The solid red line shows the calculated magnon frequency spectrum $\omega_{q_{\parallel}}$ which has two minima $\omega_{\text{bot}} \equiv \min_{q_{\parallel}} \{\omega_{q_{\parallel}}\} \approx 4 \times (2\pi) \text{ GHz}$ at $q_{\parallel} = \pm q_{\text{bot}}$ with $q_{\text{bot}} \approx 4 \times 10^4 \text{ cm}^{-1}$. The two brightest spots in the vicinity of the bottom of the magnon spectra at $\pm q_{\text{bot}}$ originate from “bottom” magnons associated with the left and right BEC states.

Above these brightest spots we see three spots with $\omega \approx 2\omega_{\text{bot}}$ and $q_{\text{left}} \approx -2q_{\text{bot}}$, $q_{\text{center}} \approx 0$, and $q_{\text{right}} \approx 2q_{\text{bot}}$. It is natural to relate them to the confluence of two bottom magnons [38,39], as shown by green arrows in Fig. 1(a): (i) left spot, $\omega_{-q_{\parallel}} + \omega_{-q_{\parallel}} \Rightarrow 2\omega_{\text{bot}}$ and $q = -2q_{\text{bot}}$; (ii) central spot, $\omega_{-q_{\parallel}} + \omega_{+q_{\parallel}} \Rightarrow 2\omega_{\text{bot}}$ and $q = 0$; and (iii) right spot, $\omega_{+q_{\parallel}} + \omega_{+q_{\parallel}} \Rightarrow 2\omega_{\text{bot}}$ and $q = 2q_{\text{bot}}$. However, in our magnetization geometry, neither $\omega = 2\omega_{\text{bot}}$ with $q = \pm 2q_{\text{bot}}$ nor $\omega = 2\omega_{\text{bot}}$ with $q = 0$ are eigenmodes of the YIG film chosen. The only plausible explanation [40] builds on the fact that we observe off-resonant waves driven by appropriate nonlinearity, i.e., virtual magnons. For brevity, we will call them “virtual double-bottom” magnons [41].

In Fig. 1(a) the BLS spectra $I(\omega, q_{\parallel})$ are supplemented by two down-pointing orange arrows showing the process of parametric pumping by an external quasihomogeneous microwave field with wave vector $\mathbf{q}_{\text{pump}} \approx 0$ and frequency ω_{pump} . Precisely at this position, we see a rather bright spot, indicating the presence of precession with this wave vector and frequency. Since there is no corresponding magnon eigenmode with $\omega = \omega_{\text{pump}}$, we observe here one more type of virtual magnon, called, for concreteness, “virtual pump” magnons. They are directly driven by the external microwave signal through the transverse component of the oscillating magnetic field [23,42,43]. The driving mechanism is similar to the excitation of ferromagnetic resonance (FMR), with the difference that in our experiment the frequency of the excitation force exceeds the FMR frequency by more than 6 GHz, while the FMR linewidth is about 1.5 MHz.

At the same time, we see no BLS response at the frequency of the parametrically pumped real magnons $\omega_{\text{par}} = \omega_{\text{pump}}/2$ because the wave numbers of the parametric magnons are quite large and lie outside the sensitivity limit

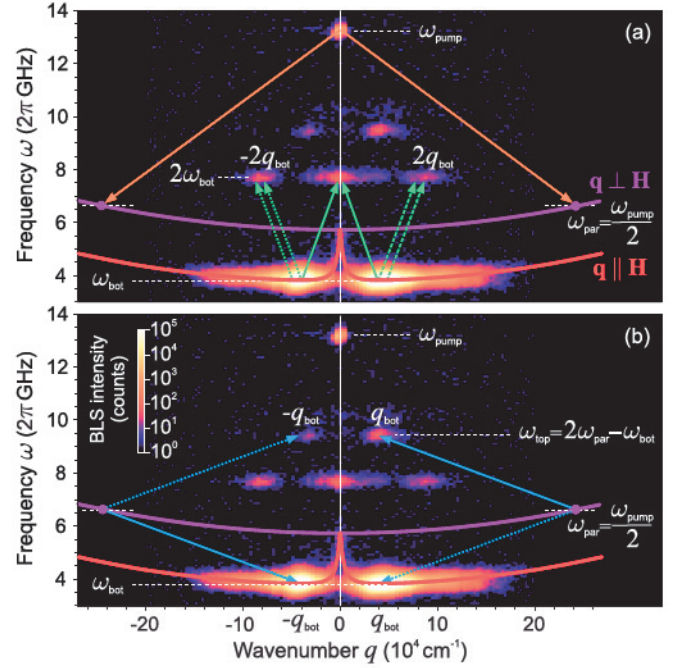


FIG. 1. Frequency- and wave-vector-resolved BLS intensity spectra of real and virtual magnons. The spectra were measured during the action of microwave pumping in a YIG film magnetized in plane by the field $H = 1350 \text{ Oe}$. The experimental intensity spectra are shown together with the calculated magnon dispersion curves [35,36] and diagrams representing the relevant quasiparticle scattering processes in the system. (a) The process of parametric pumping is shown by orange arrows. The signal of “virtual pump” magnons is visible at the pumping frequency ω_{pump} . The parametrically excited real magnons, marked by magenta dots, are outside the observation region. “Virtual double-bottom” magnons at the frequency $2\omega_{\text{bot}}$ arise due to the confluence of “bottom” magnons at ω_{bot} . The three types of confluence processes are indicated by pairs of green arrows. (b) Processes of four-wave interaction of bottom magnons with phase-correlated pairs of parametric magnons leading to the appearance of “virtual top” magnons at the frequency ω_{top} are pairwise shown by the solid and dotted blue arrows [37].

of our BLS setup at $\approx 2 \times 10^5 \text{ cm}^{-1}$. Nevertheless, these magnons are definitely present in the system [31], being responsible for the appearance of magnons at the bottom of the frequency spectrum [25,44], which are clearly visible as extremely intense BLS signals.

In our previous studies [45–48], the magnon gas thermalization and BEC formation were developing in close interactions with the phonon bath [46,49,50], and leading to accumulation of hybrid magnon-phonon quasiparticles. In the present experiments, we observe no such phenomenon because the width of the pumping area is small, and the strong efflux of these quasiparticles leads to large losses for them [45].

Two more spots at $\omega_{\text{top}} = \omega_{\text{pump}} - \omega_{\text{bot}}$ and $q_{\text{top}} = \pm q_{\text{bot}}$, have a more sophisticated origin, which we discuss below within the classical Hamiltonian

formalism convenient for description of nonlinear processes at large occupation numbers.

The classical Hamiltonian formalism is based on the equation of motion for the complex canonical wave amplitudes a_q and a_q^* , which are classical limits of the Bose creation and annihilation operators $\hat{a}_q$ and $\hat{a}_q^\dagger$ [26,27,29]:

$$\frac{da_q(t)}{dt} = -i \frac{\delta \mathcal{H}}{\delta a_q^*(t)}, \quad (1)$$

where $\mathcal{H}$ is the Hamiltonian function, called for brevity the Hamiltonian. As shown, for example, in Refs. [36,51] the classical Landau-Lifshitz equation [52] governing the motion of the magnetic momentum $\mathbf{M}(\mathbf{r}, t)$ can be represented in the Hamiltonian form (1) using the classical analog of the Holstein-Primakoff transformation from the spin operators to the Bose operators.

For weakly interacting waves, $\mathcal{H}$ can be expanded in a_q and a_q^* as $\mathcal{H} = \mathcal{H}_2 + \mathcal{H}_{\text{int}}$. The quadratic Hamiltonian $\mathcal{H}_2 = \sum_q \omega_q a_q a_q^*$ describes the free propagation of noninteracting waves with the dispersion law ω_q : $a_q(t) \propto \exp[-i\omega_q t]$. The interaction Hamiltonian $\mathcal{H}_{\text{int}} = \mathcal{H}_3 + \mathcal{H}_4$, includes

$$\mathcal{H}_3 = \frac{1}{2} \sum_{1,2,3} \left[V_{1,2}^3 a_1 a_2 a_3^* + (V_{1,2}^3)^* a_1^* a_2^* a_3 \right] \Delta_{1,2}^3, \quad (2a)$$

$$\mathcal{H}_4 = \frac{1}{4} \sum_{1,2,3,4} T_{1,2}^{3,4} a_1 a_2 a_3^* a_4^* \Delta_{1,2}^{3,4}. \quad (2b)$$

Here, $V_{1,2}^3$ and $T_{1,2}^{3,4}$ are three- and four-wave interaction amplitudes. The Kronecker symbols $\Delta_{1,2}^3$ and $\Delta_{1,2}^{3,4}$ are equal to 1 if the sum of “superscript” wave vectors equals the sum of “subscript” wave vectors, and zero otherwise, taking care of wave vector conservation. For shortness, here we use the notation $\mathbf{j} \equiv \mathbf{q}_j$, with $j = 1, 2, 3, 4$.

The close analogy between the classical Hamiltonian and a quantum mechanical description allows for a physically transparent interpretation of the interactions $\mathcal{H}_3$ and $\mathcal{H}_4$. Namely, $\mathcal{H}_3$ describes the process of the confluence of two waves (or two quasiparticles, such as magnons, phonons, etc.) with the wave vectors $\mathbf{q}_1$ and $\mathbf{q}_2$ into one wave with the wave vector $\mathbf{q}_3 = \mathbf{q}_1 + \mathbf{q}_2$, two magnons $\Rightarrow$ one magnon [see Fig. 2(a)], and the inverse process of decay of the $\mathbf{q}_3$ wave into a pair of $\mathbf{q}_1$ waves and $\mathbf{q}_2$ waves, one magnon $\Rightarrow$ two magnons [see Fig. 2(b)]:

$$\omega_1 + \omega_2 \Leftrightarrow \omega_3. \quad (3)$$

Hamiltonian $\mathcal{H}_4$ is responsible for the four-magnon scattering (two magnons $\Leftrightarrow$ two magnons) [see Fig. 2(c)] with the conservation laws

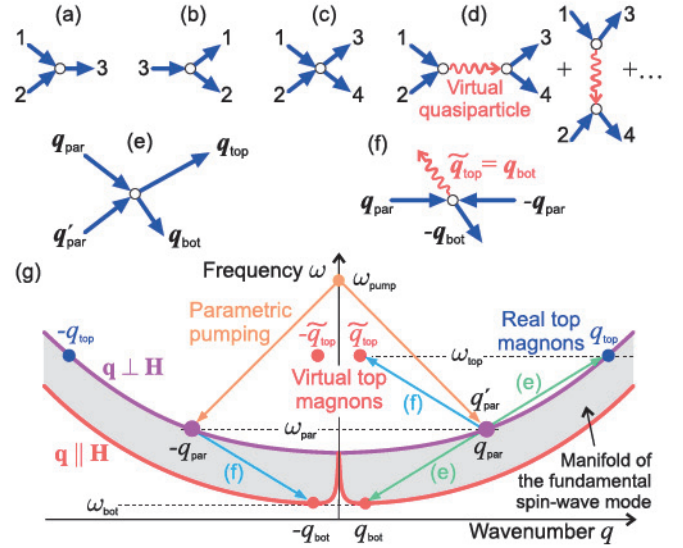


FIG. 2. Graphical representation of multiwave scattering processes. Three-wave confluence (a) and splitting (b) processes given by Eq. (3). (c) The four-wave scattering process given by Eq. (4). (d) The first (see Ref. [53]) and second contribution to the four-magnon scattering process in the second-order perturbation approach in the three-wave Hamiltonian, Eq. (2a). The red wavy lines show virtual quasiparticles, mediating these processes. All six contributions can be found, for example, in Eq. (1.1.32) from Ref. [27]. (e) Diagram of the four-magnon scattering [Eq. (7)] leading to the kinetic instability of real magnons. (f) Diagram of the four-magnon scattering [Eq. (7)] that leads to the excitation of the virtual top magnons. (g) The scattering processes shown in panels (e) and (f) are schematically represented in the magnon spectrum by pairs of green and blue arrows [54].

$$\omega_1 + \omega_2 \Leftrightarrow \omega_3 + \omega_4, \quad q_1 + q_2 = q_3 + q_4. \quad (4)$$

As a general rule, the three-magnon processes (two $\Leftrightarrow$ one) are more efficient [26]. When they are forbidden by the conservation law (3), one has to account for weaker scattering processes (two $\Leftrightarrow$ two) [Eq. (4)]. Nevertheless, three-wave processes cannot be ignored in this case either. To see this, we examine Eq. (1) with the Hamiltonian (2) away from resonances. The latter allows us not to take into account the wave damping:

$$\begin{aligned} \left[\frac{d}{dt} + i\omega_q \right] a_q = & -\frac{i}{2} \sum_{1,2} V_{1,2}^q \Delta_{1,2}^q a_1 a_2 \\ & -i \sum_{2,3} (V_{q,2}^3)^* \Delta_{q,2}^3 a_2^* a_3 \\ & -\frac{i}{2} \sum_{1,2,3} T_{1,2}^{3,q} a_1 a_2 a_3^* \Delta_{1,2}^{3,q}. \end{aligned} \quad (5)$$

The first term in the right-hand side of Eq. (5) is the sum $\sum_{1,2}$ of off-resonant forces F_{12} , each having the wave vector $\mathbf{q}_1 + \mathbf{q}_2$ and the frequency $\omega_1 + \omega_2$. Each partial

force F_{12} stirs up a wave with wave vector $\mathbf{q}_1 + \mathbf{q}_2$, the eigenfrequency $\omega_{12} \equiv \omega(\mathbf{q}_1 + \mathbf{q}_2)$, and amplitude a_{12} . Far away from resonance, $a_{12} \propto F_{12}/(\omega_1 + \omega_2 - \omega_{12})$ and, therefore, the total system response is given by the following expression:

$$\sum_{1,2} a_{12} = \sum_{1,2} V_{1,2}^{1+2} a_1 a_2 [2(\omega_1 + \omega_2 - \omega_{12})]^{-1}. \quad (6)$$

Taking a_1 and a_2 as $a_{\pm q_{\parallel}}$, we find the amplitudes of the virtual double magnons discussed above. One can say that the interaction process of $\mathbf{q}_1$ quasiparticles and $\mathbf{q}_2$ quasiparticles nonresonantly excites a wave perturbation of a nonlinear medium—virtual quasiparticles with a total amplitude given by Eq. (6). Note that these virtual quasiparticles can decay into two real quasiparticles $\mathbf{q}_3$ and $\mathbf{q}_4$ with frequencies ω_3 and ω_4 , resulting in the effective four-wave scattering process [53] schematically shown in Fig. 2(d).

Returning to the analysis of our experimental results, note that in our geometry and for $H = 1350$ Oe the high population of the bottom magnons is the result of the so-called kinetic instability [25,38,55]: a four-magnon process (4) of decay of parametric magnons with frequency $\omega_1 = \omega_2 = \omega_{\text{par}} = \omega_{\text{pump}}/2$ and wave vectors $\mathbf{q}_1 = \mathbf{q}_{\text{par}}$, and $\mathbf{q}_2 = \mathbf{q}'_{\text{par}}$ into a pair of the bottom and the so-called “top” magnons with frequencies ω_{bot} and ω_{top} [see Fig. 2(e)]. Note that $\mathbf{q}_{\text{par}}$ does not necessarily belong to the same pair of parametrically generated magnons for which $\mathbf{q}_{\text{par}} + \mathbf{q}'_{\text{par}} = 0$. If so, one expects

$$\omega_1 + \omega_2 = \omega_{\text{bot}} + \omega_{\text{top}} = \omega_{\text{pump}}, \quad \text{and} \quad (7a)$$

$$\mathbf{q}_1 + \mathbf{q}_2 = \mathbf{q}_{\text{bot}} + \mathbf{q}_{\text{top}}. \quad (7b)$$

Assuming a rough estimate that $\omega_{\text{top}} \gg \omega_{\text{bot}}$, we conclude that, as shown in Fig. 2(g) with two blue circles, $q_{\text{top}} > q_{\text{par}}$. It means that the top magnons lie outside the sensitivity

region of our BLS setup, i.e., they are invisible in Fig. 1. This raises the crucial question: What is the origin of the two spots in Fig. 1(b) at the “correct” frequency $\omega_{\text{top}} = \omega_{\text{pump}} - \omega_{\text{bot}}$, which is consistent with Eq. (7a), but with the experimentally observed wave vectors $\tilde{\mathbf{q}}_{\text{top}} = \pm \mathbf{q}_{\text{bot}}$, which completely disagree with Eq. (7b)?

To resolve this controversy, we note that the theory of kinetic instability is formulated in the framework of the weak-wave kinetic equation, which assumes weak correlations of the wave phases. As a result, the scattering (4) of real magnons has a stochastic nature and appears only as a second-order perturbation of the four-wave interaction amplitudes $T_{1,2}^{3,4}$ [Eq. (2b)]. Nevertheless, in our particular case with a large population of parametric magnons, the scattering waves have strong externally determined phase correlations. As shown in the S -theory [27], the energy flux from an external spatial homogeneous pumping field to parametric magnons is proportional to $\langle a_{\mathbf{q}_{\text{par}}} a_{-\mathbf{q}_{\text{par}}} \exp(i\omega_{\text{pump}} t) \rangle$. As a result, in the competition of parametric pairs, only the pairs with the maximum energy flux from the pump field survive, i.e., pairs with the full phase correlation, for which $|\langle a_{\mathbf{q}_{\text{par}}} a_{-\mathbf{q}_{\text{par}}} \exp(i\omega_{\text{pump}} t) \rangle| = \langle |a_{\mathbf{q}_{\text{par}}}|^2 \rangle \equiv n_{\mathbf{q}_{\text{par}}}$. This allows us to consider a pair of parametric magnons ($a_{\mathbf{q}_{\text{par}}} a_{-\mathbf{q}_{\text{par}}}$) as a “single” wave object with the frequency $2\omega_{\text{par}} = \omega_{\text{pump}}$ and phase being the sum of the phases of the waves composing the pair. Therefore, owing to its dynamic nature, the four-wave scattering process (4) with $\mathbf{q}_1 = -\mathbf{q}_2$, $\mathbf{q}_3 = \mp \mathbf{q}_{\text{bot}}$, and $\mathbf{q}_4 = \tilde{\mathbf{q}}_{\text{top}} = \pm \mathbf{q}_{\text{bot}}$ is much stronger than stochastic scattering (4) with $\mathbf{q}_1 \neq -\mathbf{q}_2$, being now proportional to the first power of the interaction amplitude $T_{1,2}^{3,4}$. One can see this from the last term in the right-hand side of Eq. (5), which describes a driving force

$$F_{\text{virt}}(\omega, \pm \mathbf{q}_{\text{bot}}) = \sum_{\mathbf{q}_{\text{par}}} T_{\mathbf{q}_{\text{par}}, -\mathbf{q}_{\text{par}}}^{\mp \mathbf{q}_{\text{bot}}, \pm \mathbf{q}_{\text{bot}}} a_{\mathbf{q}_{\text{bot}}}^* a_{\mathbf{q}_{\text{par}}} a_{-\mathbf{q}_{\text{par}}} \quad (8)$$

having the same frequency $\omega_{\text{top}} = \omega_{\text{pump}} - \omega_{\text{bot}}$ as that of real top magnons [see Eq. (7a)] and wave vector $\tilde{\mathbf{q}}_{\text{top}} = \pm \mathbf{q}_{\text{bot}}$.

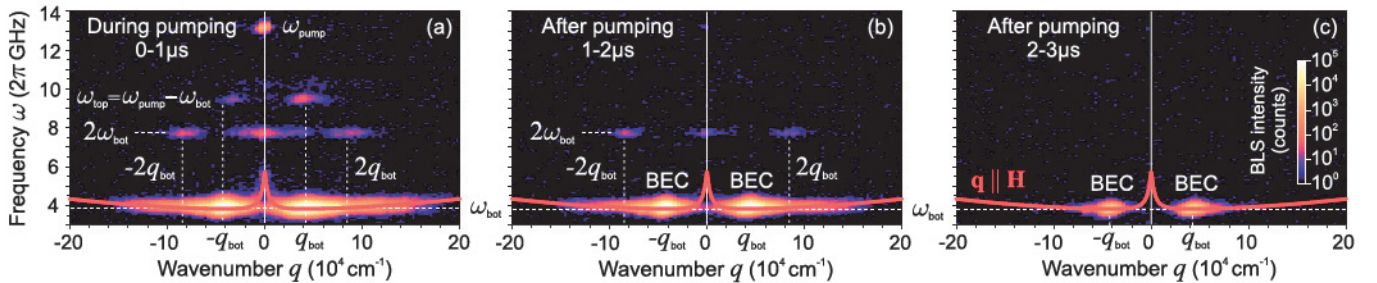


FIG. 3. Time evolution of the BLS spectra of the real and virtual magnons. (a) During the pumping microwave pulse, the virtual top magnons at $\omega_{\text{top}} = \omega_{\text{pump}} - \omega_{\text{bot}}$ are visible alongside with double-bottom virtual magnons at $2\omega_{\text{bot}}$. (b) Soon after the pumping pulse, the virtual top magnons disappear, and only double-bottom virtual magnons are visible as the populations of magnon BECs at ω_{bot} are still large. (c) 2 μs after the pumping pulse, only magnon BECs are visible. Owing to the small magnon quantity, virtual magnons are not observable. All panels: the red solid curves show the calculated dispersion curves of magnons with uniform distributions of dynamic magnetization over the thickness of the YIG film and propagating in the film plane along the magnetization field H .

This force should off-resonantly excite “virtual top” magnons [see Figs. 2(f) and 2(g)] with amplitude

$$\tilde{a}_{\text{top}}(\omega_{\text{top}}, \pm \mathbf{q}_{\text{bot}}) = F_{\text{virt}}(\omega, \pm \mathbf{q}_{\text{bot}}) / [2(\omega_{\text{bot}} - \omega_{\text{par}})]. \quad (9)$$

They are visible in Fig. 1 at $\omega = \omega_{\text{top}}$ [56].

It is worth noting that such an enhancement of the efficiency of nonlinear processes in the case of phase-correlated waves is not a specific feature of the magnon system. For example, it is known in quantum optics for simpler cases when only a few waves are involved in the interaction process [57].

To further confirm the origin of different groups of virtual magnons, we studied their evolution after the pumping is turned off. Figure 3 presents the BLS spectra of the real and virtual magnons at different moments of time. Figure 3(a) shows spectra during the pumping process (same as presented in Fig. 1, for reference). Figure 3(b) refers to the time interval $t = (1 - 2) \mu\text{s}$ after the pumping power has been turned off and the rapidly relaxing parametric magnons have disappeared. Therefore, we do not see the pump and top virtual magnons, forced either directly by the pumping magnetic field or through the parametric magnons, as seen in Fig. 2(f). The signal of the top virtual magnons disappears in a time not exceeding 20 ns. This value correlates well with earlier measurements of the scattering time of a group of parametrically pumped magnons, which in the BEC experiments was close to 10 ns [23]. Nevertheless, the double-bottom virtual magnons are still there, forced only by the bottom real magnons. They disappear only later [see Fig. 3(c)], when the number of the (real) bottom magnons becomes too small to force the double magnons above a measurable level. Thus, the time evolution of the virtual magnons confirms the mechanism of their excitation suggested in our Letter.

Traditionally, nonlinear-wave interactions are investigated via observation of real quasiparticles—quanta of freely propagating waves, i.e., eigenmodes of the nonlinear medium. In our experiments with highly populated magnon gases in a ferromagnetic YIG film, we found several types of virtual magnons—quanta of off-resonant spin waves—driven by real magnons through the appropriate nonlinearity. We show that virtual quasiparticles (virtual magnons in our case) may be the end result of a new type of nonlinear processes in systems with narrow intense packets of quasiparticles. In particular, we discovered a four-wave interaction involving pairs of parametric waves with opposite wave vectors and a magnon Bose-Einstein condensate. This process is strongly enhanced by phase correlation in parametric pairs and BECs. The considered interactions that lead to the appearance of virtual quasiparticles contribute to the behavior of nonlinear systems, and are useful for the identification of physical processes in them. Furthermore, the study of various types of virtual and

real quasiparticles and their correlation-enhanced interactions is necessary both in the classical [58,59] and quantum [58,60–62] limits, for example, for their application in new information transfer and data processing technologies, including quantum magnonics [58,60–62] and coherent quantum optics (see, e.g., Ref. [57] and references therein).

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