Thermalization of a Parametrically Driven Magnon Gas Leading to Bose-Einstein Condensation

V. E. Demidov, O. Dzyapko, and S. O. Demokritov*
Institute for Applied Physics and Center for Nonlinear Science, University of Münster, Germany

G. A. Melkov
Department of Radiophysics, National Taras Schevchenko University of Kiev, Kiev, Ukraine

A. N. Slavin
Department of Physics, Oakland University, Rochester, Michigan, USA
(Received 19 January 2007; published 20 July 2007)

The thermalization of parametrically pumped magnons caused by nonlinear multimagnon scattering processes and leading to the magnon Bose-Einstein condensation is investigated experimentally with high temporal resolution. The threshold pumping power necessary for the thermalization is determined. For pumping powers above this threshold the thermalization time has been found to decrease rapidly with power reaching the value down to 50 ns, which is much smaller than the magnon lifetime.

DOI: 10.1103/PhysRevLett.99.037205 PACS numbers: 75.30.Ds, 03.75.Nt, 76.60.Es

The relaxation of elementary magnetic excitations—magnons—in magnetically ordered substances has been intensively studied since the observation of ferromagnetic resonance (see, e.g., [1] and chapters 12–14 in [2]). Recent discoveries of precessional switching [3] and the spin-torque effects of a dc current [4] initiated an additional interest to the problem supported by possible applications of these new effects in the magnetic information storage technology.

Despite a large number of publications on the topic, the main part of the experimental data on the magnon relaxation is related only to the properties of *primary* magnons created in linear ferromagnetic resonance (FMR) or parametric pumping (see, e.g., [2]). In these experiments the information about the primary magnon relaxation was extracted from the FMR linewidth or from the threshold of parametric excitation. The *secondary* magnons, created as products of magnon-magnon relaxation of the primary magnons, were either not considered at all, or were considered as a thermal bath characterized by a given temperature and by the zero chemical potential [5].

The development of the space- and time-resolved Brillouin light scattering technique for thin magnetic film samples [6] brought an opportunity for direct studies of multimagnon relaxation processes in a dense gas of secondary parametric magnons. In particular, recent experiments [7] have demonstrated that the relaxation processes in the system of secondary parametric magnons can change the thermodynamic properties of the magnon gas in such a way that the gas undergoes Bose-Einstein condensation (BEC). For this fundamental quantum transition thermalization of the secondary magnons apparently plays a critical role.

In this Letter we report on the experimental study of the thermalization processes of parametrically excited magnons, their influence on the entire gas of magnons, and the onset of Bose-Einstein condensation. Our experiments, being performed with the high temporal resolution, allowed us to observe directly how the primary magnons created by parametric pumping relax, and how the secondary magnons created in this nonlinear relaxation process redistribute over the magnon spectrum.

Bose-Einstein condensation of quasiparticles such as excitons, biexcitons, polaritons, phonons, and magnons has recently attracted enormous attention (see, e.g., [7–19]). One of the main advantages of quasiparticles (in comparison with atoms) is their small mass, which results in BEC of quasiparticles at relatively high temperatures. Observation of BEC in a gas of magnons [7] has shown that a phase transition to a condensate can even be achieved at room temperature if the density of magnons is high enough. At the same time, the possibility of Bose-Einstein condensation of quasiparticles in the thermodynamic sense is not evident [20] since quasiparticles are characterized by a finite lifetime, which is often comparable to the time a system needs to reach thermal equilibrium. Therefore, the study of the thermalization processes for a gas of quasiparticles is of a special importance for the clear understanding of the phase transition observed in [7].

In this Letter we show that the thermalization of magnons represents the gradual population of magnon states with progressively decreasing frequencies, starting from the states close to the frequency of the primary pumped magnons and ending with the state at the bottom of the magnon spectrum where BEC of magnons takes place [7]. This fact clearly shows that the relaxation of primary magnons into the magnons with smaller energies happens through the multiple magnon-magnon scattering events, and, therefore, the magnons reaching the bottom of the spectrum lose the initial phase coherence, which might be introduced by the external pumping source. In addition, we clearly demonstrate that the speed of the thermalization depends on the density of the injected magnons, i.e., on the pumping power. We show that with the increasing pumping power the thermalization time decreases quickly and reaches a value of about 50 ns. This gives a clear evidence of the applicability of the approach of the thermodynamic quasiequilibrium used in [7].

In our experiments we used a 2×2 mm^2 square sample made of an epitaxial film of yttrium iron garnet (YIG) with the thickness of 5 μm. The sample was placed into a uniform in-plane static magnetic field. The parametric pumping was realized by means of a microstrip resonator with a width of 25 μm attached to the YIG sample along its middle line. The pumping was performed with microwave pulses of the 1 μs duration with the repetition period of 11 μs at the carrier frequency of $f_P = 8.037$ GHz, causing injection of parametric primary magnons at the frequency of $f_P/2 = 4.018$ GHz. The peak power of the pumping pulses was varied from 0.1 to 1.3 W. The redistribution of magnons over the spectrum was studied with a temporal resolution of 10 ns using the time-resolved Brillouin light scattering (BLS) spectroscopy in the quasi-backward scattering geometry [6]. This technique allows one to measure the frequency spectrum of magnons having wave vectors in the range of $k = \pm 2 \times 10^5$ cm^{-1} with the minimum frequency resolution of 50 MHz [7]. The experiments were performed at room temperature.

Figure 1 shows the low-energy part of the magnon spectrum in an in-plane magnetized ferromagnetic film calculated for the parameters of the used YIG film and the magnetic field of $H = 700$ Oe. The solid lines represent the dispersion curves for the two limiting cases of magnons with wave vectors $\mathbf{k}$ oriented parallel (the so-called backward volume waves) or perpendicularly (the so-called surface waves) to the static magnetic field $\mathbf{H}$ as indicated in the Fig. 1. Both curves merge for $k = 0$ at the frequency of the uniform FMR $f_\perp = 3.66$ GHz. The magnon states for intermediate angles fill the manifold between these two boundaries. As seen from Fig. 1, the manifold is characterized by a nonzero minimum frequency $f_{\rm Min}$ corresponding to a nonzero wave vector $\mathbf{k}_{\rm Min}$ aligned parallel to the static magnetic field. For our experimental conditions they were $f_{\rm Min} \approx 2.1$ GHz and $k_{\rm Min} \approx 5 \times 10^4$ cm^{-1}, respectively. The primary magnons created by parametric pumping at a frequency of $f_P/2$ are distributed over the states with different $\mathbf{k}$ as shown in Fig. 1 by the dashed line.

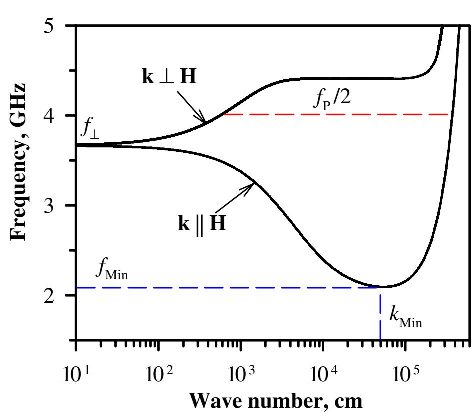

FIG. 1 (color online). Dispersion spectrum of magnons in an in-plane magnetized YIG film placed into the static magnetic field of $H = 700$ Oe. The two solid lines show the upper and the lower boundaries of the spectrum corresponding to the cases of magnons with wave vectors $\mathbf{k}$ being perpendicular and parallel to the static magnetic field $\mathbf{H}$. $f_\perp$ is the frequency of the uniform ferromagnetic resonance, $f_{\rm Min}$ is the minimum frequency of magnons at $k_{\rm Min}$, and $f_P/2$ is one half of the pumping frequency corresponding to the frequency of primary injected magnons.

In high-quality epitaxial YIG films the main mechanism responsible for the energy transfer *out* of the magnon system is the spin-lattice (magnon-phonon) interaction, which determines the typical magnon lifetime of 1 μs (see, e.g., Chap. 12 in [1]). The main processes of energy redistribution *within* the magnon system are the two- and four-magnon scattering processes (see Chap. 11 in [1]). These magnon-magnon relaxation processes lead to the spreading of injected magnons over the phase space, while keeping the number of interacting magnons in the system constant. If the magnon-magnon scattering mechanisms preserving the number of magnons are much faster than spin-lattice relaxation, the pumping creates a magnon gas characterized by a quasiequilibrium distribution of magnons over frequency (energy) spectrum.

The evolution of the magnon distribution under the influence of parametric pumping for the pumping power of 0.7 W and the static magnetic field of $H = 700$ Oe is shown in Fig. 2. This figure represents the BLS spectra accumulated for different delay times after the start of the pumping pulse. Note here, that the BLS intensity at a given frequency is proportional to the reduced density of magnons at this frequency [21], which is equal to the product of the occupation function and the reduced spectral density of magnon states. The later is obtained by integration of the magnon states over the wave vector interval accessible in the BLS experiment ($|k| < 2 \times 10^5$ cm^{-1}).

Figure 2(a) shows the reference BLS spectrum of thermally excited magnons accumulated without pumping. These magnons are at the thermal equilibrium with the crystalline lattice. They are excited and annihilated due to the interaction between the spin-system and the lattice, and their total number is defined by the temperature of the later. Consequently, this gas of magnons is described by the statistical Bose occupation function with zero chemical potential. As seen from Fig. 2(a), the BLS spectrum is limited in frequency. The low-frequency limit corresponds to the minimum magnon frequency $f_{\rm Min}$, whereas the upper-frequency limit is due to the lack of sensitivity of the experimental setup to the magnons with large wavevectors ($|k| > 2 \times 10^5$ cm^{-1}).

When the pumping is turned on additional parametric magnons are injected into the system, as illustrated by Fig. 2(b) where the BLS spectrum corresponding to the delay time $\tau = 30$ ns after the start of the pumping pulse is

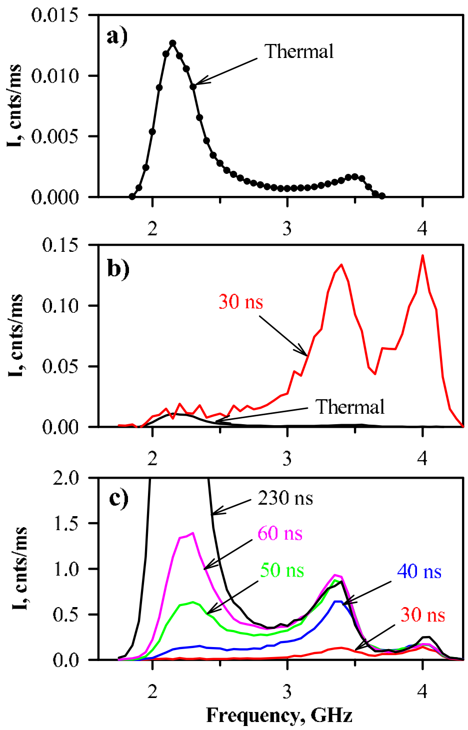

FIG. 2 (color online). The redistribution of magnons over frequencies for pumping power of 0.7 W and the static magnetic field of $H = 700$ Oe measured by means of time-resolved BLS. Shown are the frequency dependences of BLS intensity I, which is proportional to the spectral density of magnons. (a) the BLS spectrum measured without pumping, i.e., the spectrum of magnons at the thermal equilibrium with the crystalline lattice. (b)–(c) the BLS spectra measured after the pumping is turned on at delays with respect to the start of the pumping pulse as indicated.

shown. In the same figure the BLS spectrum of thermally excited magnons is shown for comparison as well. As seen from Fig. 2(b), in the early pumping stage the occupation function of magnons in the low-frequency range is not affected. On the contrary, the magnon density rises significantly at a frequency of about 4 GHz corresponding to the half of the pumping frequency and at the FMR frequency $f_{\perp} \cong 3.4$ GHz. The further evolution of the magnon distribution is presented in Fig. 2(c). The figure shows that the magnon density close to $f_{\perp}$ grows quickly and then saturates at about 60 ns after the start of the pumping pulse. In parallel, the density of magnons at lower frequencies starts to grow as well. Note, however, that for the lower frequencies the growth of the magnon population is slower and the saturation happens at larger delays. The observed process can be understood as a gradual occupation of magnon states starting from the frequency of primary magnons towards the minimum magnon frequency. This means that the increase in the population at the bottom of the spectrum takes place through the multiple inelastic scattering events. Thus, a very important intermediate conclusion can be made at this point: since the magnons close to the bottom of the spectrum are created through a series of multiple scattering events not conserving the phase of individual magnons, any coherency observed in the gas of magnons at the bottom of the spectrum must be a spontaneous one.

To determine the thermalization time in the system of secondary parametric magnons we calculated the ratio of the magnon spectra experimentally measured at different delay times after the start of the pumping pulse to the spectrum of thermal magnons measured without pumping. This ratio representing the occupation function of pumped magnons normalized by the occupation function of thermal magnons is plotted in Fig. 3; curves presented in Fig. 3 are normalized to unity at f_{Min}. The figure clearly illustrates an overpopulation of high-frequency magnon states at small delay times, $\tau = 30$–70 ns. This overpopulation, however, decreases with time and the ratio corresponding to $\tau = 230$ ns is almost constant. This fact means that the magnon distribution over frequencies at $\tau = 230$ ns is described by an equilibrium statistical occupation function with the zero chemical potential. Therefore, the time of 230 ns can be considered as the thermalization time at a given pumping power of 0.7 W.

For further illustration of the magnon dynamics the temporal dependences of the BLS intensity at two characteristic frequencies are shown in Fig. 4. The Figure shows that the occupation at the frequency of 3.4 GHz, which is far from the bottom of the spectrum, grows quickly and saturates at about 60 ns, whereas the growth of the occupation at the frequency f_{Min} clearly demonstrates two stages. In the first stage lasting about 230 ns, the occupation grows relatively fast and then saturates. This stage corresponds to the thermalization process in the magnon gas. After the thermalization is achieved a second stage characterized by a quasiadiabatic increase of the chemical potential in the thermalized magnon gas due to the growth of the total magnon density caused by the pumping starts [7].

For pumping powers smaller than 0.7 W a stationary nonthermalized distribution defined by the energy flow

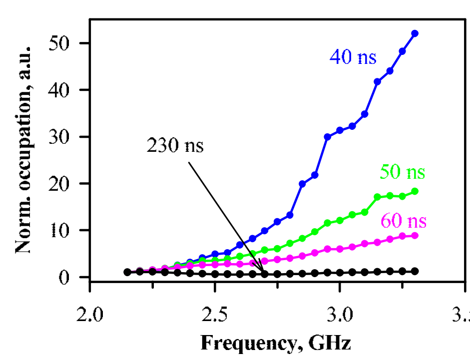

FIG. 3 (color online). The ratio between BLS spectra of pumped magnons to the BLS spectrum of thermally excited magnons at different delays with respect to the start of the pumping pulse. The experimental conditions are the same as for Fig. 2.

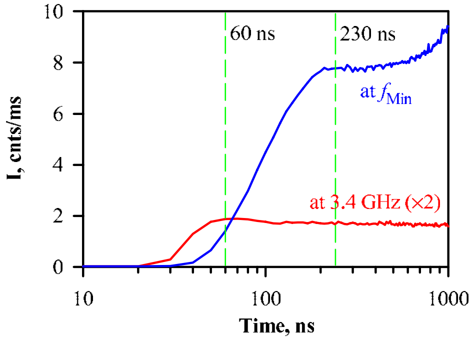

FIG. 4 (color online). The temporal dependences of the BLS intensity at two characteristic frequencies: the minimum magnon frequency $f_{Min} = 2.1$ GHz and the frequency of 3.4 GHz, which is far from the bottom of the magnon spectrum.

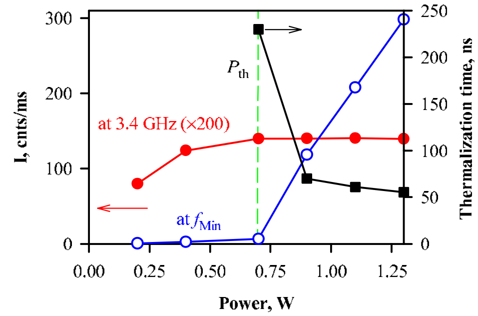

FIG. 5 (color online). Circles—dependences of the magnon densities at the characteristic frequencies f_{Min} and 3.4 GHz on the pumping power. Squares—thermalization time for pumping powers above the thermalization threshold $P_{th} = 0.7$ W.

equilibrium between the pumping and the spin-lattice relaxation is created instead of thermalization. This is explained by the fact that the intensity of the four-magnon scattering process, which is mainly responsible for the magnon thermalization, depends on the magnon density. For relatively small pumping powers (small magnon densities) the thermalization time in the magnon system is comparable to (or larger than) the magnon lifetime, and the main part of the injected magnons annihilate before they reach the bottom of the spectrum. The thermalization time rapidly decreases with power for pumping powers larger than the threshold value of 0.7 W, as illustrated by Fig. 5. As seen from the figure, the thermalization time approaches a value of about 50 ns at the pumping power of 1.3 W, which is much smaller than the lifetime of magnons in YIG films due to the spin-lattice interaction.

Figure 5 also shows BLS intensities at f_{Min} and far from it (at 3.4 GHz) versus the pumping power obtained at the delay time of 1 μs corresponding to the end of the pumping pulse. As seen from Fig. 5, the limit density of magnons far from f_{Min} grows with increasing pumping power and saturates at the power of 0.7 W, which represents the onset of the thermalization, as discussed above. On the contrary, the limit magnon density at the bottom of the spectrum experiences a strong increase above this power (note different scales for both values in Fig. 5). This result gives further support to the conclusion that as soon as the thermalization in the magnon gas is achieved, its chemical potential starts to grow, and the magnon occupation changes mainly at the frequencies that are close to the bottom of the spectrum.

In conclusion, we report on the experimental investigations of thermalization in a parametrically driven gas of magnons. The obtained results clearly show that the thermalization of magnons injected by means of parametric pumping occurs in a series of multiple scattering events not conserving the phase of individual magnons. Therefore, magnons in the prepared gas are incoherent, despite the fact that the gas is driven by a coherent external electromagnetic signal.

Support by the Deutsche Forschungsgemeinschaft, U. S. Army Research Office (MURI Grant No. W911NF-04-1-0247), and by the Science & Technology Center of Ukraine is gratefully acknowledged.

*Corresponding author.
demokrit@uni-muenster.de

[1] M. Sparks, *Ferromagnetic Relaxation Theory* (McGraw-Hill, New York, 1964).
[2] A. G. Gurevich and G. A. Melkov, *Magnetization Oscillations and Waves* (CRC, New York, 1996).
[3] C. H. Back *et al.*, Science **285**, 864 (1999).
[4] E. B. Myers *et al.*, Science **285**, 867 (1999).
[5] A. I. Akhiezer, W. G. Bar'yakhtar, and S. W. Peletminsky, *Spin Waves* (Wiley, New York, 1968).
[6] S. O. Demokritov, B. Hillebrands, and A. N. Slavin, Phys. Rep. **348**, 441 (2001).
[7] S. O. Demokritov *et al.*, Nature (London) **443**, 430 (2006).
[8] T. Fukuzawa, E. E. Mendez, and J. M. Hong, Phys. Rev. Lett. **64**, 3066 (1990).
[9] Y. Yamamoto, Nature (London) **405**, 629 (2000).
[10] T. Nikuni, M. Oshikawa, A. Oosawa, and H. Tanaka, Phys. Rev. Lett. **84**, 5868 (2000).
[11] R. Coldea *et al.*, Phys. Rev. Lett. **88**, 137203 (2002).
[12] Ch. Rüegg *et al.*, Nature (London) **423**, 62 (2003).
[13] L. V. Butov, J. Phys. Condens. Matter **16**, R1577 (2004).
[14] J. P. Eisenstein and A. H. MacDonald, Nature (London) **432**, 691 (2004).
[15] O. V. Misochko, M. Hase, K. Ishioka, and M. Kitajima, Phys. Lett. A **321**, 381 (2004).
[16] M. Jaime *et al.*, Phys. Rev. Lett. **93**, 087203 (2004).
[17] T. Radu *et al.*, Phys. Rev. Lett. **95**, 127202 (2005).
[18] Y. D. Kalafati and V. L. Safonov, Sov. Phys. JETP **68**, 1162 (1989).
[19] J. Kasprzak *et al.*, Nature (London) **443**, 409 (2006).
[20] D. Snoke, Nature (London) **443**, 403 (2006).
[21] M. Cottam and D. Lockwood, *Light Scattering in Magnetic Solids* (Wiley, New York, 1986).