Quantum coherence due to Bose–Einstein condensation of parametrically driven magnons

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Abstract. The room-temperature kinetics and thermodynamics of the magnon gas driven by microwave pumping has been investigated by means of the Brillouin light scattering (BLS) technique. We show that for high enough pumping powers the quantum relaxation of the driven gas results in a quasi-equilibrium state described by the Bose–Einstein statistics with a nonzero chemical potential. Further increase of the pumping power causes a Bose–Einstein condensation in the magnon gas documented by an observation of the magnon accumulation at the lowest energy level. Using the sensitivity of the BLS to the coherence degree of the scattering magnons, we confirm the spontaneous emergence of coherence of the magnons accumulated at the bottom of the spectrum, if their density exceeds a critical value.

The origin of the ferromagnetic state is the quantum-mechanical exchange interaction between spins. The transition paramagnet–ferromagnet is documented by a divergence of the statistical correlation length describing the correlation between spins located far from each other. The fluctuations above the ground state of a ferromagnet are usually described by means of quantized low-energy spin-wave excitations, which are magnons. Magnons at thermal equilibrium do not usually show coherence effects. In fact, they are considered to form a gas of excitations, nicely described by the quantum representation of population numbers. There have been attempts

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to describe coherent magnon states [1] by analogy to coherent photon states [2]. However, this description has not been well developed and has not been widely used yet for analysis of experimental results. Up to now, very few examples of coherent magnons can be found: ferromagnetic resonance is one of them. However, the coherence in this case is not spontaneous, but induced by an external microwave field.

One of the most striking quantum phenomena leading to spontaneous quantum coherence on the macroscopic scale is Bose–Einstein condensation (BEC). It describes the formation of a collective quantum state of bosons. As the temperature of the boson gas $T$ decreases at a given density $N$ or, vice versa, the density increases at a given temperature, the chemical potential $\mu$, describing the gas, increases as well. On the other hand, $\mu$ cannot be larger than the minimum energy of the bosons $\varepsilon_{\text{min}}$. The condition $\mu(N, T) = \varepsilon_{\text{min}}$ defines a critical density $N_c(T)$. If the density of the particles in the system is larger than $N_c$, BEC takes place: the gas is spontaneously divided into two fractions: (i) incoherent particles with the density $N_c$ distributed over the entire spectrum of possible boson states; and (ii) a coherent ensemble of particles accumulated in the lowest state with $\varepsilon = \varepsilon_{\text{min}}$ [3].

At temperatures far below the temperature of magnetic ordering, $T_c$, magnons can be considered as weakly interacting bosons: the Bloch law for the temperature dependence of the static magnetization, which nicely describes a bulk amount of experimental data, has been obtained based on this assumption. Since magnons are bosons, one can expect that they undergo the BEC transition. Several groups have reported observation of the field-induced BEC of magnetic excitations in quantum antiferromagnets TiCuCl$_3$ [4, 5], Cs$_2$CuCl$_4$ [6, 7] and BaCuSi$_2$O$_6$ [8]. In these materials, a phase transition accompanied by a magnetic mode softening occurs if the applied magnetic field is strong enough to overcome the antiferromagnetic exchange coupling. Such a transition can be treated as BEC in an ensemble of magnetic bosons. However, these excitations can hardly be considered as magnons—quanta of waves of spin precession propagating in a magnetically ordered system.

Very recently, it was shown that magnons continuously driven by microwave parametric pumping can enormously overpopulate the lowest energy level even at room temperature [9]. This observation has been associated with the BEC of magnons. At the same time, the possibility of BEC of quasi-particles in the thermodynamic sense is not evident [10] since quasi-particles are characterized by a finite lifetime, which is often comparable to the time a system needs to reach thermal equilibrium. Moreover, an observation of spontaneous coherence is an important proof for the existence of BEC. Therefore, the study of the thermalization processes for a gas of magnons and the experimental observation of the spontaneous coherence of the magnons overpopulating the lowest state are of special importance for a clear understanding of the phase transition observed in the earlier work [9].

In this paper, we present data on the thermodynamic properties of the magnon gas driven by microwave pumping, which demonstrate the above mentioned effect of the magnon accumulation. Besides, very recent results on the kinetics of the magnon gas under conditions of parametric pumping are presented. Finally, an experimental study of the magnon gas pumped by very short intensive pumping pulses and relaxing after the pumping has been switched off, clearly demonstrating the creation of a coherent magnon condensate from incoherent magnons with increasing magnon density, is discussed. All these findings together bring direct evidence of BEC of magnons.

The experiments on room temperature BEC of magnons were performed on monocrystalline films of yttrium iron garnet (YIG) with the thickness of 5 $\mu$m. YIG
$\text{Y}_3\text{Fe}_2(\text{FeO}_4)_3$ is one of the most studied magnetic substances. YIG films are characterized by very small magnetic losses providing a long magnon lifetime in this substance: it appears to be much longer than the characteristic time of magnon–magnon interaction \cite{11, 12}. This relation is a necessary precondition for the BEC in a gas of quasi-particles whose number is not exactly conserved \cite{10}. Samples with lateral sizes of several millimeters were cut from the films and were placed into a uniform static magnetic field of $H = 700–1000$ Oe oriented in the plane of the film. The injection of magnons was performed by means of parallel parametric pumping with a frequency of 8.0–8.1 GHz. The pumping field was created using a microstrip resonator with a width of 25 $\mu$m attached to the surface of the sample. The peak pumping power was varied from 0.1 to 6 W. Two types of pumping pulses were used: (i) long pumping pulses of 1 $\mu$s duration; and (ii) short pumping pulses with the duration of 30 ns, which is shorter than the characteristic thermalization time in the magnon gas \cite{11}. In the first case of long pumping pulses, the magnon dynamics was investigated during the pulse, whereas using short pumping pulses and studying the system after the pulse one has the possibility to follow the dynamics of a magnon gas that is free from any external driving force. Details of the pumping process can be found in \cite{9, 11, 13, 14}.

The redistribution of magnons over the spectrum was studied with a temporal resolution of 10 ns using time-resolved Brillouin light scattering (BLS) spectroscopy in quasi-backward scattering geometry \cite{15}. In this geometry, magnons from the wavevector interval $k = \pm 2 \times 10^5$ cm$^{-1}$, determined by the wavevector of the incident light, contribute to BLS spectra. Thus, the BLS intensity at a given frequency is the product of the magnon population at this frequency and the reduced magnon density of states, taking into account the above wavevector interval only. Giving access to the temporal evolution of the magnon distribution, the BLS technique allows one to study the kinetics and thermodynamics of magnons. A typical frequency resolution of the experimental setup limited by the resolution of the optical spectrometer was 250 MHz. It was also possible to achieve a better resolution of $\Delta f = 50$ MHz, albeit at the expense of sensitivity \cite{9}. The experiments were performed at room temperature. A detailed description of the BLS setup used can be found elsewhere \cite{9, 13, 15}.

Figure 1 illustrates the low-energy part of the dispersion spectrum of magnons in an in-plane magnetized ferromagnetic film calculated for the parameters of the YIG film used and the magnetic field of $H = 700$ Oe. The solid lines represent the dispersion curves for the two limiting cases of magnons with wavevectors $k$ oriented parallel (the so-called backward volume waves) or perpendicularly (the so-called surface waves) to the static magnetic field $H$, as indicated in figure 1. Both the curves merge for $k = 0$ at the frequency of the uniform ferromagnetic resonance. The magnon states for intermediate angles fill the manifold between these two boundaries. As seen from figure 1, the manifold is characterized by a nonzero minimum frequency $f_{\text{min}} = 2.10$ GHz corresponding to a nonzero wavevector $k_{\text{min}} = 5 \times 10^4$ cm$^{-1}$ aligned parallel to the static magnetic field. The frequency minimum at a nonzero wavevector results from competition between the magnetic dipole interaction and the exchange interaction. Note that a change of the static magnetic field shifts $f_{\text{min}}$, whereas by changing the film thickness one varies the corresponding wavevector.

Figure 1 also illustrates the process of parametric pumping of the magnon gas. This process can be considered as a creation of two primary magnons by a microwave photon of the pumping field. It does not define the values of the magnon wavevectors. The only condition is that the two created magnons have opposite wavevectors. The pumping initiates a strongly non-equilibrium magnon distribution: a very high density of primary magnons $10^{18}–10^{19}$ cm$^{-3}$ is
Figure 1. The low-frequency part of the dispersion spectrum of magnons in a YIG film magnetized by an in-plane static magnetic field \( H = 700 \text{ Oe} \). The arrows illustrate the process of parametric pumping.

created close to the frequency \( f_p \). Although the primary magnons are excited by coherent pumping, they are not coherent with each other: two magnons are excited simultaneously, and only the sum of their phases, but not the phase of each magnon, is locked to the phase of the microwave photon.

Due to the intense magnon–magnon interaction, the primary magnons are redistributed over the phase space. The main mechanisms responsible for the energy redistribution within the magnon system are the two-magnon and the four-magnon scattering processes (see chapter 11 in [12]). Four-magnon scattering dominates in high-quality epitaxial YIG films. It can be considered as an inelastic scattering mechanism, since it changes the energies of the scattering magnons. As a consequence, the four-magnon scattering leads to the spreading of the magnons over the spectrum, keeping, however, the number of magnons in the system constant. Note here that the three-magnon scattering process, which does not conserve the number of magnons, does not play an important role in the described experiments [13]. In parallel, an energy transfer out of the magnon system due to the spin–lattice (magnon–phonon) interaction takes place. It will be shown below that the magnon–magnon scattering mechanisms preserving the number of magnons are much faster than the spin–lattice relaxation. Under these conditions a step-like pumping should create a magnon gas characterized by a steady, quasi-equilibrium distribution of magnons over the phase space after a certain transition period characterized by a thermalization time.

The magnon distributions illustrating the evolution of the magnon gas to the quasi-equilibrium state are shown in figure 2 for the pumping power of 0.7 W. This figure presents BLS spectra recorded for different delay times after the start of the pumping pulse. At the delay time \( t = 0 \) (no magnons are pumped yet) the magnon distribution corresponds to thermally excited magnons. In the early pumping stage \( (t = 30 \text{ ns}) \) the population of magnon states close to \( f_{\text{min}} \) is not affected at all. In contrast, the magnon density at frequencies from about 2.5 to 4 GHz (the latter is close to the frequency of the primary magnons) rises significantly. Further evolution of the magnon distribution presented in figure 2 shows a saturation of the magnon
Figure 2. Evolution of the magnon population after a step-like pumping has been switched on. Note a wave-like increase of the magnon population propagating from higher frequencies toward the bottom of the spectrum.

population. In fact, at $t = 60$ ns, the magnon population of the entire spectrum except the region close to $f_{\text{min}}$ is saturated. The density of magnons close to $f_{\text{min}}$ starts to grow for $t > 30$ ns and saturates for very large delays as shown in figure 3. The observed process can be understood as a gradual wave-like population of magnon states starting from the frequency of primary magnons toward the minimum magnon frequency. This means that the increase in 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 $f_{\text{min}}$ are created through a series of multiple scattering events not conserving the phase of individual magnons, any coherence observed in the gas of magnons at the bottom of the spectrum must be spontaneous.

After the magnon population at the bottom of the spectrum saturates, the entire magnon gas reaches a steady state. To check whether this steady state corresponds to a quasi-equilibrium thermodynamic state, one should determine whether it is described by the Bose–Einstein statistics. That can be done directly by fitting the measured magnon spectra recorded for large delay times. However, a more apparent way to demonstrate that the gas of secondary magnons has reached quasi-equilibrium is illustrated in figure 4, where the ratio of the magnon spectra experimentally recorded at different delay times to the spectrum of thermal magnons measured without pumping is shown. In this ratio, the influence of all additional factors (magnon density of states, accessible interval of wavevectors, etc) is compensated. This ratio represents just the population function of pumped magnons normalized by the population function of the thermal magnons. For better comparison, the curves presented in figure 4 are normalized to unity at $f_{\text{min}}$. The figure clearly illustrates a non-equilibrium overpopulation of high-frequency magnon states at small delay times, $t = 30–70$ ns, which has already been seen in figure 2. This overpopulation, however, decreases with time and the ratio corresponding to $t = 230$ ns is almost constant.
Figure 3. Evolution of the magnon population at different frequencies as a function of the delay time after a step-like pumping has been switched on. Note a slow increase in the population for $f_{\text{min}}$.

Figure 4. The ratio of the population of pumped magnons to that of thermally excited magnons at different delays with respect to the start of the pumping pulse.

This fact means that the magnon distribution over frequencies at $t = 230$ ns is described by an equilibrium statistical population function with 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. Note here, that for higher pumping powers the thermalization can happen at nonzero chemical potentials. For pumping powers smaller than 0.7 W, a stationary non-thermalized distribution defined by the energy flow equilibrium between the pumping and the spin–lattice relaxation is found. The absence of the thermalization at low powers is due to the nonlinear nature of the

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Figure 5. The thermalization time as a function of the pumping power. The shadowed area corresponds to the power below the thermalization threshold $P_{\text{th}} = 0.7$ W, where the thermalization cannot be achieved. The solid line is a guide for the eye.

Due to nonlinearity of the four-magnon scattering, the thermalization time also rapidly decreases with pumping power for its values above the threshold 0.7 W, as illustrated by figure 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 significantly smaller than the lifetime of magnons in YIG films due to the spin–lattice interaction. The shadowed area in the figure indicates the region of smaller pumping powers where the thermalization cannot be achieved.

As described above, the BEC takes place if the chemical potential reaches the value of the minimum energy of magnons. After thermal quasi-equilibrium is reached, further pumping increases the density of magnons as a function of time. As a result, the value of the chemical potential increases as well. For the used values of the pumping powers, this growth of $\mu$ occurs at a much lower rate than the thermalization process; therefore it can be considered as being adiabatic.

Figures 6(a) and (b) show the measured BLS spectra at large delay times reflecting the quasi-equilibrium distributions of magnons over the phase space at different pumping powers $P = 4$ and 5.9 W, correspondingly. Tokens in the figures represent the experimental data; solid lines are the distributions calculated based on the Bose–Einstein statistics [13], using $\mu$ as the fit parameter. As seen in figure 6(a), the chemical potential grows with time, reaching saturation at $\mu/h = 2.08$ GHz. This value is close but still below $f_{\text{min}}$. Apparently higher values of $\mu$ cannot be reached at this pumping power, since the pumped magnons leave the magnon gas due to spin–lattice relaxation. Figure 6(b) illustrates the process at $P = 5.9$ W. For this pumping power, the maximum value $\mu/h = 2.10$ GHz is already reached after 300 ns. One can conclude
Figure 6. (a) BLS spectra from pumped magnons at a pumping power of 4 W at different delay times, as indicated. Solid lines show the results of the fit of the spectra based on the Bose–Einstein statistics with the chemical potential being a fit parameter. Note that the critical value of the chemical potential cannot be reached at the power used. (b) Same as (a) but for a pumping power of 5.9 W. The critical value of the chemical potential is reached at 300 ns.

that the critical density of the magnon gas $N_c$ is achieved at $t = 300$, and the corresponding distribution can be considered as the critical distribution $n_C(f)$. Further pumping leads to a phenomenon which can indeed be interpreted as BEC of magnons: additionally pumped magnons are collected at the bottom of the spectrum without changing the population of the states with higher frequencies. The latter fact is demonstrated by figure 6(b) as well, showing the high-frequency parts of the magnon distribution curves on an appropriate scale. These data demonstrate that the BLS spectra for $t > 300$ ns cannot be described just by increasing the temperature in the Bose–Einstein population function, since a higher temperature means higher magnon populations at all frequencies. Thus, figure 6(b) indicates the formation of a Bose–Einstein condensate of magnons.

One can make out a difference between a distribution at a given time $t > 300$ ns and the critical one. One sees from figure 6(b) that this difference is nonzero just in the region close to $f_{\text{min}}$, the width of the region $\Delta f \approx 0.2–0.3$ GHz being defined by the resolution of the spectrometer. Optical measurements with the ultimate resolution have shown that the intrinsic width of the region is even below 50 MHz. Moreover, microwave spectroscopy indicates that it is narrower than 6 MHz, which corresponds to a high degree of coherence of magnons in the condensate, giving $\Delta f < 10^{-6} k_B T / h$. Thus, the narrowing of the magnon distribution with respect to that determined by the classical Boltzmann statistics is more than six orders of magnitude!
Figure 7. Magnon density for the primary magnons (blue tokens) and magnons at the bottom of the dispersion spectrum (red tokens). The dashed line presents the result of an exponential fit as discussed in the text. From the latter curve, the magnon lifetime is determined.

However, the question of quantum coherence is not trivial, especially if one takes into account that the system under investigation is a quasi-equilibrium one. In fact, due to finite lifetime of the particles, each quantum state is spread in frequency, the width of the spreading $\Delta f$ being determined by the lifetime, $\tau_0$: $\Delta f \approx (2\pi \tau_0)^{-1}$. To study the magnon system, which is free from any influence of the external driving force and to determine the magnon lifetime, the width of pumping pulses has been changed: instead of 1 $\mu$s long pulses, as for all previously considered data, very short pulses of 30 ns duration were used for pumping, as will be discussed below. Note here that for new experimental conditions, a new stripe resonator has been developed; therefore a direct comparison of the pumping powers provided above and below is impossible. The experiments with short pumping pulses were performed at the applied magnetic field of 1000 Oe, which causes an increase in $f_{\text{min}}$ to 3.0 GHz.

In a similar way as shown in figure 3 for long pumping pulses, figure 7 shows the evolution of the magnon density at $f_P$ and $f_{\text{min}}$ for $P = 2$ W. It demonstrates that the density of primary magnons rises very fast and then decreases due to four-magnon scattering which distributes the pumped magnons over the phase space. The dynamics of the magnon population at $f_{\text{min}}$ is rather different: it takes about 200 ns before the density starts to grow reaching its maximum at $t = 550$ ns and then it decreases exponentially. The lifetime $\tau_0 = 260$ ns determined from the exponential part of the curve is due to the energy transfer into the lattice caused by the spin-lattice relaxation. Note here that this time is much longer than the magnon–magnon scattering time at high pumping powers (cf figure 5). Therefore, one can consider the magnon gas as a quasi-equilibrium system.
Figure 8. Color representation of the BLS intensity portraying the population of pumped magnons as a function of the frequency and the delay time with respect to the start of the pumping pulse for different pumping powers.

The process of relaxation of the magnon gas is further illustrated in figure 8, where two-dimensional maps presenting the normalized BLS intensities as a function of the magnon frequency and the delay time are shown for different pumping powers. First, it is seen from the figure that the redistribution of the primary magnons over the phase space happens faster for larger pumping powers. This fact is direct evidence of the nonlinear nature of the four-magnon scattering. More surprising is the fact that the decay of the BLS intensity at $f_{\text{min}}$ apparently depends on the power. This very surprising fact is further illustrated in figure 9, where the temporal dependences of the BLS intensity are shown for different pumping powers corresponding to different numbers of magnons injected during the pumping pulse. The data are shown on a logarithmic scale. Since the absolute value of the scattering intensity strongly depends on the pumping power, the data shown here are normalized at $t = 1000$ ns. The dashed lines represent the results of the best fit of the experimental points by the exponential decay function. Since the formation of the overpopulated state at $f_{\text{min}}$ happens faster for higher pumping powers, the data for smaller powers start at larger delays.

To understand the shown effect, one should consider the relation between the BLS intensity and the magnon density in more detail. The BLS intensity is proportional to the temporal average of the square of the electric field of the scattered light, $E$ (see e.g. [16]). In the case of many scatterers the total scattering intensity is proportional to $\langle (\sum E_i)^2 \rangle$, where $E_i$ is the scattered field of the $i$th scatterer. It is obvious that the total scattering intensity of incoherent scatterers is proportional to $\langle \sum E_i^2 \rangle$, i.e. to the number of scatterers, $N$, due to zeroing of all cross-correlators of scattered fields: $\langle E_i E_j \rangle = 0$, $i \neq j$. In contrast, in the case of in-phase coherent scatterers with equal scattering amplitudes, the above cross-correlators are not zero $\langle E_i E_j \rangle = \langle E_i^2 \rangle$ and the scattering intensity is proportional to $N^2$. Thus, comparing the temporal dependence of the BLS intensity and that of the number of scattering magnons, one can detect...
Figure 9. Decay of the BLS intensity at the lowest magnon frequency $f_{\text{min}}$ after the formation of the overpopulated state is finished for different values of the pumping power, as indicated. Lines—best fit of the experimental points by the exponential decay function. The inset shows an example of the fit of the measured magnon distributions based on the Bose–Einstein statistics.

the magnon coherence. In fact, if the magnon density decays $n \propto \exp(-t/\tau_0)$, the BLS intensity in the case of incoherent magnons should follow the same function. However, for the case of coherent magnons, the BLS intensity should follow $n^2$: $(\exp(-t/\tau_0))^2 = \exp(-2t/\tau_0) = \exp(-t/(\tau_0/2))$. In other words, for the same decay of the magnon density, the BLS intensity decays twice as fast for coherent magnons than for incoherent magnons. The experimentally measured decay times of the BLS intensity at $f_{\text{min}}$ as a function of the pumping power $P$ are presented in figure 10. It is clear from figure 10 that the decay time reduces in a stepwise way its value by a factor of two with pumping power increasing from 2.5 to 4 W. Based on the above analysis, one has to conclude that for pumping powers $P \geq 2.5$ W, the magnons at $f_{\text{min}}$ start to form a coherent Bose–Einstein condensate and the contribution of the condensate to BLS dominates for $P \geq 4$ W. This fact is further corroborated by the inset in figure 9, where the magnon distribution for the maximum used pumping power $P = 6$ W and $t = 200$ ns is shown together with the results of the fit using the Bose–Einstein distribution function. One can see that Bose–Einstein statistics nicely describes the magnon distribution in the entire frequency range except the point $f_{\text{min}}$ where a narrow peak (note the logarithmic scale) is created indicating the existence of a magnon condensate. Thus, our experimental data clearly show that the magnons accumulated at $f_{\text{min}}$ are coherent at high enough pumping powers and this coherence emerges spontaneously if the density of the magnons exceeds a certain critical value. This finding gives direct experimental evidence of the BEC transition in a magnon gas at room temperature.

The BEC transition and emergence of coherence in the magnon gas should also lead to an abrupt increase of the BLS intensity at $f_{\text{min}}$, since $N^2 \gg N$. In order to demonstrate this fact,
Figure 10. Blue circles—dependence of the measured decay time of the BLS intensity at the lowest magnon frequency $f_{\text{min}}$ as a function of the pumping power. Corresponding line—fit of the experimental data with the sigmoid function. Red circles—the maximum detected BLS intensity at the frequency $f_{\text{min}}$ versus the pumping power. Corresponding line—a guide for the eye.

the maximum value of the BLS intensity at $f_{\text{min}}$ versus pumping power $P$ is plotted in figure 10 as well. One should pay attention to the logarithmic scale for the y-axis and to the fact that the experimental error for this curve is smaller than the size of the tokens. As seen from the figure, the dependence demonstrates a clear kink at $P = 2.5$ W, marking the onset of the observed BEC transition. Although the quantitative relation between the measured BLS intensity and the true order parameter for the transition (density of the condensate) is not clear at the moment, one can conclude that a phase transition characterized by a continuous change of the order parameter at the transition point is observed. Such behavior is typical, e.g., for the superfluid transition in liquid helium or superconductivity in metals.

In conclusion, we have investigated the thermalization of magnon gas driven by microwave parametric pumping to a quasi-equilibrium state with a nonzero chemical potential. For a certain critical value of the pumping power, the BEC of magnons occurs. For short pumping pulses, this state is free from any influence of the external driving force and is mainly governed by the internal interactions between magnons. We found that starting from a certain critical density, the accumulated magnons become coherent. The obtained results are in accordance with the concept of BEC and give undoubted experimental evidence of the existence of a Bose–Einstein condensate at room temperature.

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