Crossover behavior in the phase transition of the Bose-Einstein condensation in a microwave-driven magnon gas

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(Received 1 August 2009; published 30 September 2009)

A magnon gas in a film of yttrium iron garnet driven by microwave radiation exhibits Bose-Einstein condensation (BEC) when the driving power exceeds a critical value. We show that the nature and the critical exponents of the BEC transition change dramatically if the BEC magnons are significantly coupled to the zone-center magnons. The theoretical results explain the diverse behavior of the order parameter inferred from the experimental data for the light scattering and the microwave emission from the BEC observed with coherent and incoherent microwave pumping.

DOI: 10.1103/PhysRevB.80.092409 PACS number(s): 75.30.Ds, 03.75.Nt, 75.45.+j

It has been well-established experimentally[1–5] and theoretically[6] that quasiequilibrium Bose-Einstein condensation (BEC) of magnons is achieved at room temperature in films of yttrium-iron garnet (YIG) driven by microwave radiation if the driving power exceeds a critical value. The BEC state is characterized by coherence, by a number of condensate magnons close to the number of pumped magnons and by an order parameter represented by a macroscopic small-signal transverse magnetization.[6] The properties of the BEC have been observed and measured experimentally with Brillouin light scattering (BLS) techniques[1–3] and through the microwave emission from uniform magnons generated by BEC magnon pairs.[4,5]

In a YIG film magnetized by an applied in-plane field, the combined effects of the exchange and magnetic dipolar interactions among the spins produce a magnon dispersion relation (frequency ω_k versus wave vector k) that has a minimum ω_{k0} at $k_0 \sim 10^5$ cm^{-1}. The dynamics of magnons driven parametrically in the film by a pulsed microwave magnetic field has been studied by Brillouin light scattering with time, frequency, and wave-vector resolution.[1–3] The experiments show that if the power of the pumping microwave with frequency $f_p=8.1$ GHz exceeds a first threshold value, there is a large increase in the population of the parametric magnons with frequency in a narrow range around $f_p/2=4.05$ GHz. Then the energy of these primary magnons redistributes in about 50 ns through modes with lower frequencies down to the minimum frequency $f_{\min}=\omega_{k0}/2\pi=2.9$ GHz (for $H=1.0$ kOe) as a result of magnon interactions that conserve the number of magnons. This produces a hot magnon gas that remains decoupled from the lattice for several hundred ns due to the long spin-lattice relaxation time. As the microwave power is increased the chemical potential rises and approaches the minimum energy producing an overpopulation of magnons around that frequency.

The BLS experiments reveal that if the microwave power exceeds a second threshold value, much larger than the one for parallel pumping, the magnon population condenses in a narrow region in phase space around the minimum frequency $f_{\min}$ and develops quantum coherence.[2,3] The coherence of the magnon condensate is also demonstrated in another set of experiments[4,5] in which the applied in-plane static field has a value such that the frequency of the $k\approx 0$ magnon is $\omega_0=2\omega_{k0}$. In this case a microwave radiation signal is generated by $k\approx 0$ magnons created by pairs of BEC magnons k_0, $-k_0$ through a three-magnon confluent process. The $k\approx 0$ value is necessary for emission because the wave number of electromagnetic radiation with frequency $f=1.5$ GHz is $k=2\pi f/c \approx 0.3$ cm^{-1}. Microwave emission is observed whether the microwave driving is coherent[4] or incoherent,[5] demonstrating the coherence of the BEC magnons. In an earlier paper[6] we presented a theoretical model for the dynamics of magnons in a YIG film driven by microwave radiation far out of equilibrium that provides rigorous support for the formation of Bose-Einstein condensation of magnons. The model relies on the cooperative action of magnons with frequencies close to $f_{\min}$ through the nonlinear four-magnon interactions.

The theory provides the basic requirements for the characterization of a BEC, namely: (a) the onset of the BEC is characterized by a phase transition that takes place as the microwave power p is increased and exceeds a critical value p_{c2}; (b) the magnons in the condensate are in coherent states and as such they have nonzero small-signal transverse magnetization that is the order parameter of the BEC; (c) for $p > p_{c2}$ the magnon system separates in two parts, one in thermal equilibrium with the reservoir and one with N_0 coherent magnons having frequencies and wave vectors in a very narrow region of phase space. As the microwave power increases further N_0 approaches the total number of magnons pumped into the system characterizing unequivocally a Bose-Einstein condensation. In this paper we show that the resonant coupling of BEC magnon pairs to $k\approx 0$ magnons affects considerably the dynamics of the formation of the magnon BEC. As a result the nature and the critical exponents of the BEC phase transition change dramatically if the BEC magnons are significantly coupled to the zone-center magnons. The theoretical results explain the diverse behavior of the order parameter inferred from the experimental data for the light scattering and the microwave emission from the BEC magnons.

We use a quantum spin-wave formalism to treat the excitations of a magnetic system described by the following Hamiltonian,

$$H = H_0 + H^{(3)} + H^{(4)} + H'_{eff}(t), \tag{1}$$

where

$$H_0 = \hbar \sum_k \omega_k c_k^+ c_k \tag{2}$$

is the unperturbed Hamiltonian representing free magnons with frequency ω_k and wave vector $\vec{k}$ described by creation and annihilation operators c_k^+ and c_k, $H^{(3)}$ and $H^{(4)}$ represent the nonlinear magnetic interactions and $H'_{eff}(t)$ the effective driving of the $\vec{k}_0$, $-\vec{k}_0$ magnon pairs through the collective action of the magnon gas created by the microwave pumping.[6] The magnetic Hamiltonian is dominated by Zeeman, exchange, and magnetic dipolar contributions. For small values of the wave number such that the wavelength is comparable to the shortest sample dimension the magnon frequency depends strongly on the dipolar interaction and this produces the minimum in the dispersion relation away from the zone center.

As is well known the eigenstates $|n_k\rangle$ of the free Hamiltonian H_0 and of the number operator $n_k = c_k^+ c_k$ can obtained by applying integral powers of the creation operator to the vacuum. These states have precisely defined number of magnons n_k, but they have uncertain phase and zero expectation value for the small-signal transverse magnetization operators m_x and m_y, where $\hat{z}$ is the equilibrium direction of the magnetization $\vec{M} = \hat{z}M_z + \hat{x}m_x + \hat{y}m_y$. Thus the eigenstates of n_k do not have a macroscopic wave function. The states that correspond to classical spin waves are coherent magnon states[7,8] defined as the eigenkets of the circularly polarized magnetization operator $m^+ = m_x + im_y$. They can be written as the direct product of single-mode coherent states, the eigenstates of the annihilation operator, $c_k|\alpha_k\rangle = \alpha_k|\alpha_k\rangle$, the eigenvalue α_k being a complex number. The coherent states do not have well-defined number of magnons but they have nonzero expectation values for the magnetization m^+ with a well-defined phase. It can be shown[7–9] that the coherent state $|\alpha_k\rangle$ has an expectation value for the number operator given by $\langle n_k\rangle = |\alpha_k|^2$ and a small-signal transverse magnetization

$$m^+ \propto |\alpha_k| = \langle n_k\rangle^{1/2}. \tag{3}$$

In Hamiltonian (1) the only terms in $H^{(3)}$ relevant for the dynamics considered here are the ones describing the three-magnon confluence process[10,11]

$$H^{(3)} = \hbar V_{(3)} c_0^+ c_{k0} c_{-k0} + \text{H.c.}, \tag{4}$$

where the vertex of the interaction for small wave vectors is dominated by the dipolar interaction and is given approximately by $V_{(3)} = \omega_M/(2SN)^{1/2}$, N being the number of spins S, $\omega_M = \gamma 4\pi M$, where M is the saturation magnetization and $\gamma = g\mu_B/\hbar$ is the gyromagnetic ratio (2.8 GHz/kOe for YIG). In the process studied here the term $H^{(4)}$ in Eq. (1) representing the four-magnon interaction has contributions conserving energy and momentum given by[10–12]

$$H^{(4)} = \hbar \sum_{k,k'} \left(\frac{1}{2} S_{kk'} c_k^+ c_{-k}^+ c_{k'} c_{-k'} + T_{kk'} c_k^+ c_{k'}^+ c_k c_{k'} \right), \tag{5}$$

where the interaction coefficients are determined mainly by the dipolar and exchange energies. For the k values relevant to the experiments the exchange contribution is negligible so that the coefficients in Eq. (5) are given approximately by the bulk values for the dipolar interaction $S_{kk'} = 2T_{kk'} = 2\omega_M/NS$.[10] In Eq. (1) $H'_{eff}(t)$ is the Hamiltonian for driving $\vec{k}_0$, $-\vec{k}_0$ magnon pairs given by[6]

$$H'_{eff}(t) \cong \hbar (h\rho)_{eff} e^{-i2\omega_{k0}t} c_{k_0}^+ c_{-k_0}^+ + \text{H.c.}, \tag{6}$$

where

$$(h\rho)_{eff} = -i\eta_m [(p - p_{c1})/(p_{c2} - p_{c1})]^{1/2} \tag{7}$$

represents an effective driving field expressed in terms of the microwave pumping power p, the critical power p_{c2} for the formation of the BEC, the critical power p_{c1} for parallel pumping and the intermagnon relaxation rate η_m.

Using Hamiltonian (1) with Eqs. (4)–(6) as the interaction and driving terms one can write the Heisenberg equations for the operators c_k and c_k^+ from which several quantities of interest can be obtained. One of them is the correlation function σ_k defined by,[12]

$$\sigma_k = \langle c_k c_{-k}\rangle = n_k e^{i\varphi_k} e^{-i2\omega_k t}, \tag{8}$$

where n_k is the magnon number operator and φ_k is the phase between the states k and $-k$. To study the process by which the pairs of BEC coherent magnons $\vec{k}_0$, $-\vec{k}_0$ are produced and then generate $k \approx 0$ modes we use Hamiltonian (1) to obtain the equations of motion for the magnon operators c_0 and c_{k0},

$$\frac{dc_0}{dt} = -(i\omega_0 + \eta_0 + iV_{(4)}n_0)c_0 - iV_{(3)}c_{k0}c_{-k0}, \tag{9}$$

$$\frac{dc_{k0}}{dt} = -(i\omega_{k0} + \eta_{k0} + i2V_{(4)}n_{k0})c_{k0} - i[V_{(3)}c_0 + (h\rho)_{eff}e^{-i2\omega_{k0}t}]c_{-k0}^+, \tag{10}$$

where $V_{(4)} = S_{kk} + 2T_{kk} = 4\omega_M/NS$ and the relaxation rates were introduced phenomenologically. We consider that all states involved are coherent magnon states as demonstrated earlier[6,13] and work with the corresponding eigenvalues α_k. In addition we consider that many states in a very narrow region of phase space around k_0, ω_{k0} are occupied by the condensate magnons and they act together driving the $k=0$ magnons. So we introduce in the equations a factor p_{k0} that represents the number of states occupied by condensate magnons weighted relative to the k_0 mode. The equations of motion for the eigenvalue α_0 and the correlation function $\sigma_{k0} = \alpha_{k0}\alpha_{-k0}$ in a frame rotating with frequency $2\omega_{k0}$ become

$$\frac{d\alpha_0}{dt} = -i(\omega_0 - 2\omega_{k0} + V_{(4)}n_0)\alpha_0 - \eta_0\alpha_0 - ip_{k0}V_{(3)}\sigma_{k0}, \tag{11}$$

$$\frac{d\sigma_{k0}}{dt} = -2(\eta_{k0} + i2V_{(4)}n_{k0})\sigma_{k0} - i2[V_{(3)}\alpha_0/p_{k0} + (h\rho)_{eff}]n_{k0}. \tag{12}$$

Note that in Eq. (12) the term representing the coupling with the $k=0$ mode is divided by the factor p_{k0} assumed in the driving because σ_{k0} corresponds to only one pair-mode k_0. This term represents a reaction of the $k=0$ mode that influences the behavior of the BEC modes. In steady-state $d/dt=0$ Eq. (11) leads to

$$\alpha_0 = \frac{-ip_{k0}V_{(3)}}{\eta_0 + i(\omega_0 - 2\omega_{k0} + V_{(4)}n_0)}\sigma_{k0}. \qquad (13)$$

This result shows that the BEC magnon pairs drive the uniform mode as an effective microwave magnetic field by means of the three-magnon interaction. Note that there is no threshold condition in this process, BEC magnon pairs with any value of n_{k0} will create $k=0$ magnons. However, this process is effective only when the applied field H is such the resonance condition $\omega_0=2\omega_{k0}$ is satisfied. The presence of the term $iV_{(4)}n_0$ in the denominator due to the four-magnon interaction represents a detuning from the resonance condition due to the renormalization of the $k=0$ mode frequency. In fact, this term is responsible for the saturation in the growth of the $k=0$ mode amplitude with pumping power observed experimentally in Ref. 4 when a single-frequency microwave source is used in the pumping.[6,13]

Equation (13) shows that if the applied field is such that the frequencies ω_0 and $2\omega_{k0}$ are very different, $\alpha_0 \approx 0$ so that the coupling of the BEC modes to the $k=0$ mode is ineffective. In this case the solution of Eq. (12) with $d/dt=0$ is straightforward and using Eqs. (7) and (8) one obtains the steady-state population of the k_0 mode in terms of the pumping power

$$n_{k_0} = \frac{n_H}{(p_{c2}-p_{c1})^{1/2}}(p-p_{c2})^{1/2}, \qquad (14)$$

where

$$n_H \equiv \eta_m/2V_{(4)} = \eta_m NS/8\omega_M. \qquad (15)$$

Considering the factor p_{k0} representing the modes in a narrow region of phase space around k_0, ω_{k0} one can write the number of magnons in the condensate as

$$N_0 = \frac{p_{k0}n_H}{(p_{c2}-p_{c1})^{1/2}}(p-p_{c2})^{1/2}. \qquad (16)$$

Note that the factor p_{k0} is introduced in an *ad hoc* manner in the model in order to apply the one-mode calculation to the many mode problem and it is determined by the fit of the theoretical results to the experimental data. Equations (14) and (16) are valid only for $p \geq p_{c2}$. They show that the onset of the BEC is characterized by a second-order phase transition with a critical exponent $\frac{1}{2}$ for the condensate number. Correspondingly, the small-signal magnetization that represents the order parameter of the BEC has a critical exponent $\frac{1}{4}$. Figure 1(a) shows the variation in N_0 with power calculated with $p_{k0}=4.4\times10^3$ which is a value used to fit the experimental data of Refs. 2–4. Note that this value is very small compared to the number of states $N_R \sim 10^9$ calculated numerically by counting states in k space with frequencies in the range of $\omega_{k0}-\omega_p/2$.[6]

Consider now that the resonance condition $\omega_0=2\omega_{k0}$ is satisfied so that the coupling between the modes is effective. For low values of p_{k0} Eqs. (11) and (12) can be solved in the steady state with a few approximations to obtain analytical

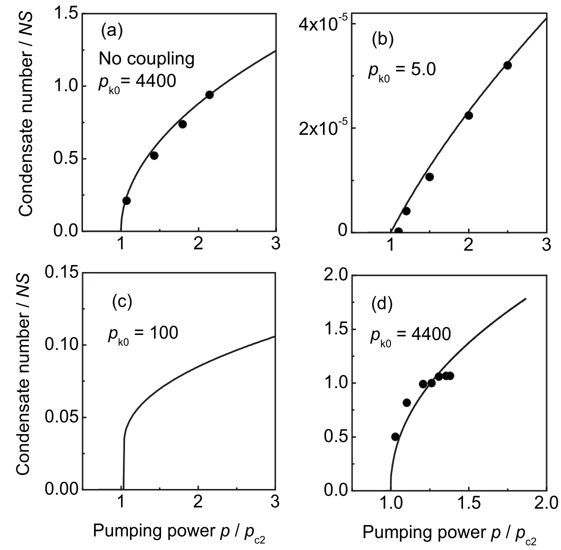

FIG. 1. Variation with microwave pumping power of the normalized steady-state magnon number of the BEC. The curves represent the theoretical results obtained with the values of p_{k0} indicated in each panel. In (a) there is no coupling between the BEC and the $k=0$ magnons while in (b), (c), and (d) there is resonant coupling with $\omega_0=2\omega_{k0}$. The symbols represent values obtained from the experimental data of Refs. 2–5 as explained in the text.

expressions for the magnon populations n_0 and n_{k0} as a function of pumping power. For low p_{k0} the number n_0 is relatively small so that $V_{(4)}n_0 \ll \eta_0$. For moderate powers above the critical value the phase of the correlation function is such that $\sigma_{k0} \approx -in_{k0}$ and Eq. (13) leads to $\alpha_0 \approx -V_{(3)}p_{k0}n_{k0}/\eta_0$. This result can be used to solve Eq. (12) with $d/dt=0$ to give the steady-state value for the population of the k_0 magnons,

$$n_{k0} = \frac{\eta_0}{V_{(3)}^2}[\,|(h\rho)_{eff}| - \eta_{k0})]. \qquad (17)$$

Note that in the presence of the coupling with the uniform mode the growth of the k_0 magnon population is limited by the three-magon interaction instead of the four magnon that prevails with no coupling. More striking is the change in the power dependence of n_{k0}. Using Eq. (7) in Eq. (17) and considering $p_{c2} \gg p_{c1}$ one obtains for pumping power near the critical value $p \geq p_{c2}$,

$$N_0 \approx \frac{p_{k0}\eta_0\eta_{k0}}{2V_{(3)}^2 p_{c2}}(p-p_{c2}). \qquad (18)$$

Equation (18) reveals that the number of magnons in the condensate varies linearly with the driving power so it has critical exponent 1 instead of $\frac{1}{2}$ as in the case of zero coupling to the $k=0$ mode. Using the relations $\alpha_0 \approx -V_{(3)}p_{k0}n_{k0}/\eta_0$ and $n_0=|\alpha_0|^2$ in Eq. (18) we obtain the power dependence of the population of the $k=0$ mode,

$$n_0 \approx \frac{p_{k0}^2\eta_{k0}^2}{4V_{(3)}^2 p_{c2}^2}(p-p_{c2})^2. \qquad (19)$$

The signal power radiated by the uniform magnetization precessing about the static field with frequency ω_0 is, as shown in Refs. 6 and 13, proportional to the number of $k \approx 0$ magnons, $p_s \propto n_0$. Thus Eq. (19) implies that for low values of p_{k0} the signal power varies quadratically with the pumping power, as observed in experiments employing incoherent microwave driving.[5]

If the number of BEC modes p_{k0} is large one cannot solve the coupled Eqs. (11) and (12) analytically. We have used a Runge Kutta code to calculate numerically the real and imaginary parts of α_0 and σ_{k0} as a function of time from which we find the steady-state values of the magnon populations n_0 and n_{k0} for each pumping power. The calculations were done assuming $\eta_0 = \eta_{k0} = \eta_m$ and using dimensionless variables and parameters: $n'_k = n_k/SN$, $t' = \eta_m t$, $V'_{(3)} = V_{(3)}(SN)^{1/2}/2\eta_m$, $V'_{(4)} = V_{(4)}(SN)/2\eta_m$ and $(h\rho)'_{eff} = (h\rho)_{eff}/\eta_m$. With $4\pi M = 1.76$ kG and $\eta_m = 5\times 10^7$ s^{-1} we have $V'_{(3)} = 219.0$ and $V'_4 = 1240.0$. The curves in panels (b), (c), and (d) in Figure 1 show the calculated variation with microwave power of the normalized steady-state number of BEC magnons for three values of p_{k0}, 5.0, 100, and 4400.

Figure 1 shows clearly the crossover behavior in the BEC phase transition with the conditions of the coupling to the $k=0$ mode. In the absence of coupling the transition is of second order and the critical exponents of the condensate number N_0 and of the small-signal magnetization of m^+ are, respectively, $\frac{1}{2}$ and $\frac{1}{4}$. For resonant coupling and low values of p_{k0} the onset of the BEC also follows a second-order phase transition, $N_0 \propto (p - p_{c2})$, $m^+ \propto (p - p_{c2})^{1/2}$, so the critical exponents are 1 and $\frac{1}{2}$. However, for resonant coupling and large values of p_{k0} the onset of the BEC is characterized by a first-order phase transition at p_{c2}.

The theoretical predictions of the model are confirmed by the experimental data of Demokritov and co-workers[1–5] obtained with two very different techniques, BLS from magnons and microwave emission from the uniform mode driven by BEC magnon pairs. The BLS experiments were carried out with applied fields such that the coupling of the BEC magnons with the $k=0$ mode is negligible. As argued in Refs. 2 and 6 the intensity of the BLS peak at the BEC frequency is proportional to the square of the condensate magnon number. The symbols in Figure 1(a) correspond to $0.026\, I_{\rm BLS}^{1/2}$, where $I_{\rm BLS}^{1/2}$ is the measured[2] BLS peak intensity as a function of power for $p \geq p_{c2} = 2.8$ W. The symbols in Figures 1(b) and 1(d) were obtained from the microwave emission data of Refs. 4 and 5, respectively, obtained in the resonant condition, considering that the signal power is $p_s \propto n_0 \propto N_0^2$. The symbols in (b) correspond to $3.45\times 10^{-5}\, p_s^{1/2}$ and in (d) to $0.473\, p_s^{1/2}$. The drastic difference between the two figures is due to the fact that in Ref. 4 the pumping is done with a single-frequency coherent microwave source whereas in Ref. 5 the pumping employs a broadband incoherent source. As shown earlier[6,13] the model fits very well the data of Ref. 4 with $p_{k0} = 4400$ and $p_{c2} = 2.8$ W. On the other hand the data of Ref. 5 are fit with $p_{k0} = 5.0$. The reason for the very large change in the fitting parameter is not quite clear. It may be caused by the change in the driving field from coherent to incoherent. But it may also be due to the fact that the experiments reported in Refs. 4 and 5 employ different spatial pumping configurations since they were carried out in different laboratories. The deviation of the data points from the theoretical curve in (d) is attributed to the fact that with coherent pumping the number of $k=0$ magnons at higher power is large and the term $iV_{(4)}n_0$ in the denominator of Eq. (13) becomes important so that p_s is not quite proportional to N_0^2.

In summary we have shown that in a YIG film driven by microwave radiation the phase transition characterizing the Bose-Einstein condensation exhibits a crossover behavior as the conditions of the coupling between the BEC and the $k=0$ magnons change. The theoretical predictions of the model are supported by the experimental data obtained with light scattering and microwave emission from the BEC magnons.

The author is grateful to Sergej Demokritov for providing important information on the experiments, to Cid B. de Araújo for many stimulating discussions, and to the Ministry of Science and Technology for supporting this work.

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