Critical parameters of nuclear magnon Bose–Einstein condensation in systems with dynamical frequency shift

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The critical conditions for the Bose–Einstein condensation of quasi equilibrium nuclear magnons in the easy-plane antiferromagnets $CsMnF_3$ and $MnCO_3$ are theoretically derived. Both systems possess the dynamical frequency shift, the dependence of the precession frequency on the magnetization deflection angle and one is able to observe the magnetic resonance signals in very non-equilibrium conditions when the frequency of pumping is higher than precession frequency. The frequency difference in this case can be compensated by the increasing of deflection angle.

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Bose–Einstein condensation (BEC) is a fabulous phenomenon of quantum statistical physics. There is a finite temperature T_c when the chemical potential of the thermodynamically equilibrium ideal gas of Bose particles reaches the minimal energy of the energy band and the particles with this energy form a coherent macroscopic quantum state [1, 2]. It was suggested that the superfluidity of ^{4}He is accompanied by BEC of atoms [3] but the strong interaction between ^{4}He atoms leads to the significant decrease of the condensate contribution even at ultra-low temperatures [4]. Theoretically predicted in 1924 BEC effect was observed in experiments with ultra cold atoms about seventy years later, in 1995 [5–7].

Magnons, Bose quasiparticles [8], elementary quantum excitations of magnetoordered systems became attractive candidate to observe BEC phenomenon at the end of eighties [9–12]. There is, however, a principal difference between Bose quasiparticles, which have finite lifetime and completely disappear at zero temperature and real Bose particles, which density is not changed at any temperature. This means that there are no conventional thermodynamic conditions for the Bose–Einstein condensation of magnons. Nevertheless, it is possible to create quasiequilibrium thermodynamic states of the higher densities and to compensate relaxation of magnons by the external pumping field. The BEC of magnons is characterized by the ordering and coherent oscillation of the macroscopic transverse magnetization. The purposeful experiments [13, 14] several years ago demonstrated BEC of magnons in parametrically pumped YIG film with a specific double-well dipolar spectrum at room temperature.

It should be noted that BEC of elementary magnetic excitations, nuclear magnons actually was observed for the first time in an exotic magnetoordered system of superfluid ^{3}He–B at ultra-low temperatures in 1984 [15, 16]. The quasiequilibrium state of nuclear magnons was created by RF pumping at the nuclear magnetic resonance (NMR) frequency and the coherent radiation from the macroscopic transverse magnetization of nuclear magnons with $k = 0$ was detected. Bose–Einstein condensation of nuclear magnons in superfluid ^{3}He was reviewed in Refs. [17–19]. In Ref. [20] it was shown that the dynamical properties of the magnetic resonance in some antiferromagnets are similar to that ones of ^{3}He-A. Later BEC of nuclear magnons with $k = 0$, but quite different physical properties than nuclear magnons in ^{3}He, was experimentally observed in the easy-plane antiferromagnets $CsMnF_3$ and $MnCO_3$ at liquid helium temperature $T = 1.5$ K [21, 22]. In this Letter we focus theoretically to non-trivial conditions of BEC of nuclear magnons in the easy-plane antiferromagnets.

Nuclear magnon is a quantum of nuclear spin wave. The concept of nuclear spin waves, collective magnetic excitations which appear in the NMR frequency range in ferro- and antiferromagnets by means of indirect Suhl–Nakamura interaction between paramag-

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netic nuclear spins, was introduced in Refs. [23, 24]. The physics of these excitations is described, for example, in Refs. [12, 25, 26]. The dispersion relation for nuclear spin waves does not depend on the direction of wave vector $\mathbf{k}$ in the easy-plane antiferromagnets and can be written as

$$\omega_{n,k} = \omega_n \sqrt{1 - \frac{H_\Delta^2(T)}{H(H+H_\mathrm{D}) + H_\Delta^2(T) + \alpha^2 k^2}}, \quad (1)$$

where ω_n is the non-shifted NMR frequency defined by the hyperfine field from the ordered electronic spin, H is the external magnetic field, H_D is the Dzyaloshinskii field, $H_\Delta^2(T) \propto 1/T$ is the gap in the electronic spin wave spectrum defined by the hyperfine field from paramagnetic nuclear spin, α is the parameter of inhomogeneous exchange interaction [27]. The difference between non-shifted NMR frequency ω_n and frequency of nuclear-electron resonance precession $\omega_L = \omega_{n,0}$ at given temperature and magnetic field is called the parameter of the dynamical frequency shift (DFS) $\omega_{p,0} = \omega_n - \omega_L$. As can be seen from Eq. (1), this parameter can be adjusted by variation of the magnetic field.

It can be seen from Fig. 1 that there are two domains of nuclear spin behaviour in k-space. At low k we

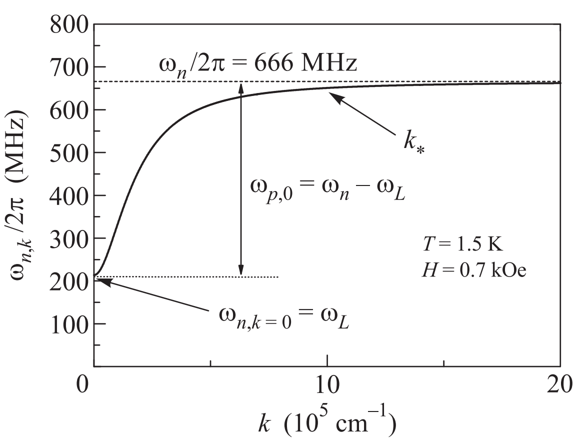

Fig. 1. The spectrum of nuclear spin waves (Eq. (1)) in antiferromagnet $\mathrm{CsMnF_3}$ ($H_\mathrm{D} = 0$, $H_\Delta^2(T) = 6.4/T\,\mathrm{kOe}^2$, $\alpha = 0.95 \cdot 10^{-5}\,\mathrm{kOe \cdot cm}$)

have the collective nuclear-electronic spin oscillations with spatial dispersion, while at high k these oscillations degenerate into a paramagnetic level. The interface $k_*(T)$ between two domains is determined by condition that the mean free path of nuclear spin wave is equal to its wavelength [28], e.g., for the case of $\mathrm{CsMnF_3}$ $k_* \sim 10^6\,\mathrm{cm}^{-1}$. Other words, calculating critical densities, $\int n_\mathbf{k}\, d^3k/(2\pi)^3$, we should limit the integral at k_* because the excitations of higher k belong to paramagnetic states. It is important to point out that the temperature and magnetization of nuclear spin systems are mainly determined by these paramagnetic states due to their highest volume in the phase space.

The analysis of interactions in the nuclear system shows that process of scattering on paramagnetic fluctuations and four-quasiparticle process are effective at $T \le 2$ K [26] and therefore conserve the total number of nuclear magnons. In addition, these processes lead to a quasiequilibrium in the externally excited nuclear magnon system with the effective chemical potential μ and effective temperature, which is assumed to have a negligible deviation from the thermal bath temperature T. Thus, the Bose–Einstein occupation number $n_\mathbf{k}$ does not have any violation and can be expressed as

$$n_\mathbf{k} = \left[\exp\left(\frac{\hbar\omega_{n,k} - \mu}{k_\mathrm{B}T}\right) - 1\right]^{-1}, \quad (2)$$

where $\hbar\omega_{n,k}$ is the magnon energy. For the case of thermal nuclear magnon density we have $\mu = 0$. The relation $n_0 \to \infty$ defines the condition BEC when the total density of thermal and pumped nuclear magnons become critical and the effective chemical potential reaches the bottom of nuclear magnon band: $\mu_c = \hbar\omega_{n,0}$. The formula for the density of nuclear magnons with the effective chemical potential μ has the form

$$\frac{N(\mu,T)}{V} \simeq \int_0^{k_*} \left[\exp\left(\frac{\hbar\omega_{n,k} - \mu}{k_\mathrm{B}T}\right) - 1\right]^{-1} \frac{4\pi k^2 dk}{(2\pi)^3}. \quad (3)$$

Here $N(\mu,T)$ is the total number of thermal and pumped nuclear magnons and V is the volume of the sample.

So far as a typical nuclear magnon "temperature" $T_n = \hbar\omega_n/k_\mathrm{B} \sim 10^{-2}\,\mathrm{K}$ is small, the occupation number (2) in the temperature range $10^{-2}\,\mathrm{K} \ll T \le 2\,\mathrm{K}$ can be written in the form

$$\left[\exp\left(\frac{\hbar\omega_{n,k} - \mu}{k_\mathrm{B}T}\right) - 1\right]^{-1} \simeq \frac{k_\mathrm{B}T}{\hbar\omega_{n,k} - \mu}. \quad (4)$$

Note that this high-temperature approximation for distribution function is valid for all nuclear magnons in their phase space $k < k_*$ and effective chemical potential $0 \le \mu < \hbar\omega_{n,0}$. This is one more unique physical property of nuclear magnons. In contrast, all other systems of Bose particles and quasiparticles demonstrate high-temperature distribution just in the visinity of point of BEC and the occupation numbers in the major part of their phase space are exponentially small.

Let us estimate the density of nuclear magnons with the effective chemical potential μ:

$$\frac{N(\mu,T)}{V} \simeq \int_0^{k_*} \frac{k_\mathrm{B}T}{\hbar\omega_{n,k} - \mu} \frac{4\pi k^2 dk}{(2\pi)^3}. \quad (5)$$

Notice that the ratio $1/(\hbar\omega_{n,k}-\mu)$ is pretty bounded and smooth function. For the highest phase weight in the vicinity of k_* it is equal to $1/(\hbar\omega_n-\mu)$. Therefore one can rewrite Eq. (5) in the form

$$\frac{N(\mu,T)}{V}\simeq\frac{k_\mathrm{B}T}{\hbar\omega_n-\mu}\int_0^{k_*}\frac{4\pi k^2dk}{(2\pi)^3}=\frac{k_\mathrm{B}T}{\hbar\omega_n-\mu}\frac{k_*^3(T)}{6\pi^2}. \quad (6)$$

This expression is valid for all chemical potentials $\mu\leq\leq\hbar\omega_n$, including the point of Bose–Einstein condensation $\mu_c=\hbar\omega_{n,0}$ (there is no singularity at $k=0$ in Eq. (5)).

Now we are able to find the critical density of pumped nuclear magnons $N_{p,c}/V$, the important parameter, which characterizes BEC in the quasi equilibrium Bose system (in contrast to T_c in the system of real Bose particles). It is defined as a difference between the density $N(\mu_c,T)/V$ of all nuclear magnons minus the initial density of thermal magnons $N(0,T)/V$:

$$\frac{N_{p,c}}{V}=\frac{N(\mu_c,T)}{V}-\frac{N(0,T)}{V}\simeq \simeq\frac{k_*^3(T)}{6\pi^2}\frac{k_\mathrm{B}T}{\hbar\left(\omega_n-\omega_{n,0}\right)}\frac{\omega_{n,0}}{\omega_n}. \quad (7)$$

Taking into account that $k_*^3(T)\propto T^{-2}$ [28], we see that the critical density of pumped nuclear magnons $N_{p,c}/V\propto 1/T$ decreases with increasing of temperature T.

It is convenient to write the ratio for the critical density of all quasi equilibrium nuclear magnons compared to the density of thermal nuclear magnons

$$\frac{N\left(\mu_c,T\right)}{N\left(0,T\right)}=\frac{\omega_n}{\omega_n-\omega_{n,0}} \quad (8)$$

and the ratio for the critical density of pumped nuclear magnons compared to the density of thermal magnons

$$\frac{N_{p,c}}{N\left(0,T\right)}=\frac{\omega_{n,0}}{\omega_n-\omega_{n,0}}. \quad (9)$$

We see that the critical number of pumped nuclear magnons should be greater than the number of thermal magnons if $\omega_{n,0}>\omega_n/2$ and smaller if $\omega_{n,0}<\omega_n/2$.

The results of the numerical calculations by Eq. (3) as well as by analytical Eq. (6) at different parameter of DFS are shown in Fig. 2. It can be seen that the approximations using for obtaining Eq. (6) are in a good agreement with the direct numerical calculations. One can see in Fig. 2 that at small DFS parameters (≤ 200 MHz) the density of condensed magnons mostly results from the pumping and the initial thermal magnons contribute less than 10 % to the critical density.

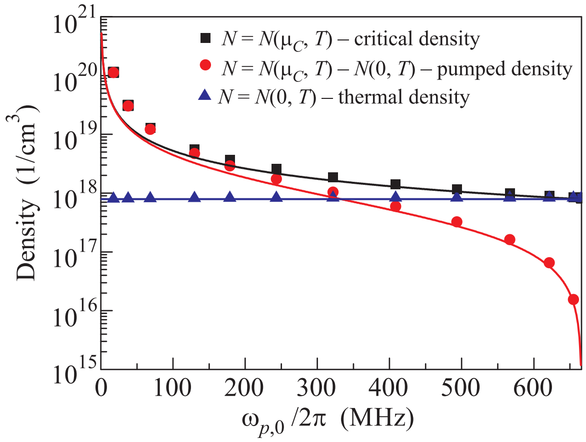

Fig. 2. (Color online) Critical density of magnons for their BEC formation, thermal magnon density and the critical density of pumped magnons as a function of parameter of DFS at temperature of 1.5 K in $CsMnF_3$. Solid curves show the dependence obtained by using Eq. (6), dots show the results of direct numerical calculations by Eq. (3)

Macroscopically the BEC state manifests itself by coherently precessing transverse magnetization with frequency equals to the bottom of the spectrum $\min(\hbar\omega_{n,k})=\omega_L$ (Fig. 1) and with amplitude

$$M_\perp=M_0\sin\beta, \quad (10)$$

where M_0 is the equilibrium nuclear magnetization, β – the magnetization deflection angle. The equilibrium magnetization is defined by the temperature and the hyperfine field as [29]

$$M_0=\frac{n\mu_n^2H_n}{3k_\mathrm{B}T}, \quad (11)$$

where n is the density of nuclei, μ_n – nuclear magnetic moment, H_n – hyperfine magnetic field.

From experimental point of view it is more conveniently to deal with deflection angles than with magnon densities, particularly when applying theory of Bose–Einstein condensation because at the energy minimum in rotating frame there is direct correspondence between pumping frequency ω_{RF} and the deflection angle [30, 31]:

$$\omega_{RF}=\omega_n-\omega_{p,0}\cos\beta. \quad (12)$$

Let us make the estimation of the critical deflection angles β_c for magnon BEC formation. The condensation of N_c non-equilibrium coherent magnons to the state with $k=0$ leads to the decreasing of the longitudinal magnetization by $\hbar\gamma N_c$:

$$M_\parallel=M_0\cos\beta_c=M_0-\hbar\gamma N_c. \quad (13)$$

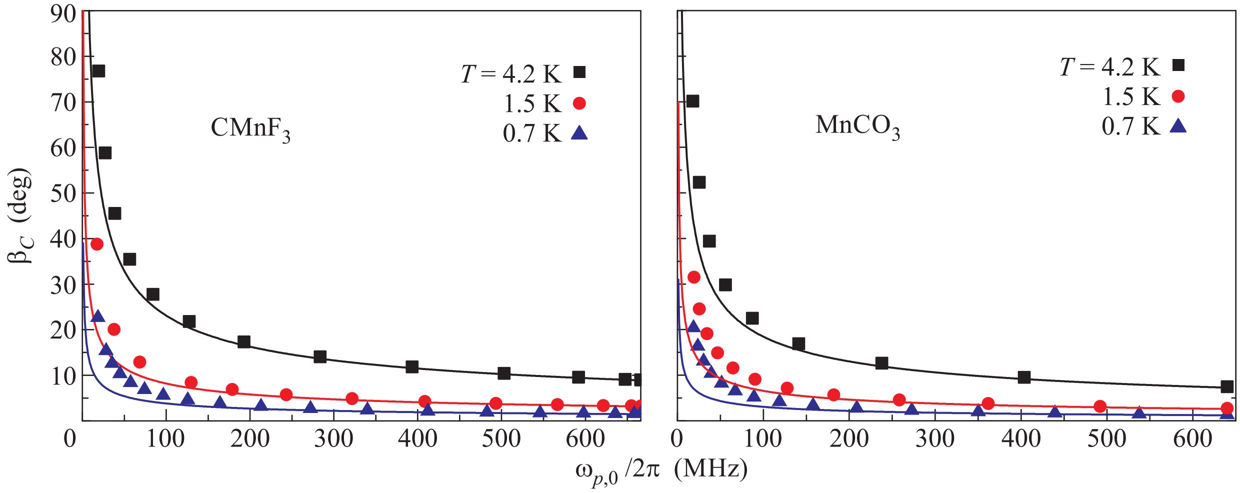

Fig. 3. (Color online) The critical deflection angle of the magnetization of the quasi-nuclear branch in $CsMnF_3$ (left panel) and in $MnCO_3$ (right panel) in dependence on the parameter of the dynamical frequency shift at different temperatures. Solid curves show the dependence obtained by using Eq. (6), dots show the results of direct numerical calculations by Eq. (3)

Eq. (13) describes the relation between the number of condensed to the $k = 0$ magnons and the magnetization deflection angle. It can be rewritten as:

$$N_c = \frac{M_0 - M_\parallel}{\hbar\gamma} = \frac{M_0}{\hbar\gamma}\left(1 - \cos\beta_c\right). \tag{14}$$

Thus, by using Eq. (3) or analytical approximation (6) at $\mu = \mu_c$ and (14) one can estimate the critical angle β_c of the magnetization deflection when BEC of nuclear magnons becomes possible. This estimation is important for choosing the optimal initial experimental conditions in further experiments with BEC of magnons. In Fig. 3 the calculations of the critical deflection angle dependence on the parameter of DFS are shown for the cases of $CsMnF_3$ (left panel) and $MnCO_3$ (right panel). It can be seen that at small values of the parameter of the dynamical frequency shift $\omega_{p,0} < 30$ MHz the critical angle is more than 10 degrees. In this case even cooling the sample down to 700 mK does not lead to the notable decreasing of the critical angle and the formation of magnon BEC is suppressed. The increasing of the parameter of DFS, that from the experimental point of view corresponds to the decreasing of the external magnetic field (see Eq. (1)), leads to the significant reduction of the critical angle. The experimental estimations of the deflection angles at magnon BEC formation were done in [32]. In those experiments the magnon BEC was formed at slow sweeping the magnetic field down (at slow increasing of the parameter of DFS). The RF pumping frequency was constant and equal to $\omega_{RF}/2\pi = 560.6$ MHz which corresponds to $\omega_{p,0}/2\pi = 105.4$ MHz. The temperature of the sample was 1.5 K. The experimental results were well described by theory of magnon BEC at deflection angles more than ~ 7 deg. It corresponds well to our calculations (Fig. 3, left panel) which show that at lower angles (when the magnon number is less than its critical value) BEC can not be formed.

In conclusion, the critical parameters of nuclear magnon BEC in the systems with the dynamical frequency shift were estimated. We believe that our theoretical results can be used for optimal choice of the experimental parameters such as temperature and magnetic field and will stimulate further experimental studies of BEC of nuclear magnons (and related phenomena) in $CsMnF_3$, $MnCO_3$, and other objects.

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