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Discovery of the Classical Bose–Einstein Condensation of Magnons in Solid Antiferromagnets

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Results of experiments in which the Bose–Einstein condensate of magnons is created in the $CsMnF_3$ easy-plane antiferromagnet in a system with coupled nuclear–electron precession with dynamical frequency shift are presented. This condensate is similar to the Bose–Einstein condensate of magnons in superfluid ^{3}He-A in aerogel.

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Bose–Einstein condensation (BEC) is one of the most striking macroscopic quantum phenomena. A macroscopic number of particles form a coherent quantum state, which is described by a common wavefunction. The formation of such a state under certain conditions was predicted by Einstein in 1925 (see review [1]). A nearly ideal BEC state has been discovered in supercooled atomic gases. The BEC state is strongly distorted in Bose liquids by intrinsic interactions, but it remains the main formation mechanism of a macroscopic quantum state. The main dynamical property of the BEC is superfluidity, which can be described as a quantum transport phenomenon emerging in the presence of BEC wavefunction gradients.

Strictly speaking, BEC theory is applicable to stable particles. However, it can also be applied to quasistable particles if their lifetime is much longer than characteristic Bose condensate formation times. In particular, although atomic gases consist of stable particles, they evaporate from a trap in about a second, but this is enough time to form the BEC. Magnons in superfluid ^{3}He are quasiparticles, but their BEC can exist for longer times [2] and it can even be maintained continuously [3].

Before we turn to the description of magnon BEC, we should clarify our terminology. A phase transition to a magnetically ordered state has been experimentally discovered in certain magnetic systems. This transition was described in terms of a change in the number of equilibrium magnons (see review [4]). Their density reached macroscopic values during the transition, indicating the formation of a new phase. This phenomenon was called the BEC of magnons. Magnons obey Bose–Einstein statistics, and appropriately, their density is described by the same formulas as those for the BEC of atoms. However, there is an important difference. In case of the classical BEC predicted by Einstein, the quantum distribution of particles changes rather than the vacuum in which they exist, whereas a phase transition is accompanied by a change in the ground state of the system (vacuum). The validity of using the term BEC to describe such states is controversial. The magnon condensate studied in this work is analogous to the atomic condensate. In order to distinguish it from a phase transition, we refer to it as a classical BEC.

The classical BEC of magnons and spin superfluidity were experimentally discovered in superfluid ^{3}He-B [5]. In subsequent studies, six different superfluid ^{3}He states in which BEC appears have been discovered. Various experiments in which the BEC was observed are reviewed in [6–8]. In all cases, the BEC forms in a system of excited nonequilibrium magnons. Pulsed or continuous pumping is used for their excitation at the NMR frequency. The magnetization becomes tilted by a certain angle β, which corresponds to the generation of magnons with the density

$$N = \frac{S - S_z}{\hbar} = \frac{\chi H}{\hbar\gamma}(1 - \cos\beta), \qquad (1)$$

where S_z is the equilibrium magnetization and S_z is its projection on the direction of the external magnetic field. Excited magnons usually precess with local precession frequencies, so that the induction signal becomes out of phase during the time of inhomogeneous broadening of the magnetic resonance line. The BEC phenomenon consists in the formation of a coherent state via magnons, where they precess with the same phase and frequency despite the inhomogeneous external conditions. In the case of continuous NMR, the pump frequency determines the chemical potential of the system. The condensate is formed within a certain region or at a low magnon density, which is determined by the chemical potential. When the NMR frequency is changed, the chemical potential also changes, which corresponds to an increase either in the condensate region or in the magnon density. There is one basic advantage as compared to the case of the atomic BEC. We can generate additional magnons, which compensate the relaxation of the coherent state, and thus the BEC can be maintained continuously. Thus, the pulse method corresponds to the formation of the BEC under a fixed particle density, while the continuous method corresponds to the formation of the BEC under a fixed chemical potential.

Theoretical description of the magnon BEC is based on the Gross–Pitaevskii equation for the order parameter:

$$\Psi(\mathbf{r}, t) = \langle \hat{\Psi}(\mathbf{r}, t) \rangle, \quad N = |\Psi|^2, \quad N = \int d^3 r |\Psi|^2, \quad (2)$$

where n is the local density of magnons. If the lifetime of magnons is long enough and their dissipation can be neglected, the Gross–Pitaevskii equation can be written in the standard form

$$-i\frac{\partial \Psi}{\partial t} = \frac{\delta \mathcal{F}}{\delta \Psi^*}, \quad (3)$$

where $\mathcal{F}\{\Psi\}$ is the free energy functional. In the case of the coherent state of precession

$$\Psi(\mathbf{r}, t) = \Psi(\mathbf{r}) e^{i\omega t} \quad (4)$$

and the Gross–Pitaevskii equation is transformed into the Ginzburg–Landau equation with $\omega = \mu$:

$$\frac{\delta \mathcal{F}}{\delta \Psi^*} - \mu \Psi = 0. \quad (5)$$

In the case of a low magnon concentration, the free energy functional in the Ginzburg–Landau theory has the form

$$\mathcal{F} - \mu \mathcal{N} = \int d^3 r \left\{ \frac{|\nabla \Psi|^2}{2m} + [\omega_L(\mathbf{r}) - \omega]|\Psi|^2 + F_{SO} \right\}, \quad (6)$$

where $\omega_L(\mathbf{r}) = \gamma H(\mathbf{r})$ is the local Larmor frequency, which plays the role of an external potential like Earth's gravitation field in the case of the atomic condensate. The last, nonlinear, term $F_{SO}(|\Psi|^2)$ appears due to spin–orbit coupling. It is analogous to the fourth-order term, which takes into account interaction in the atomic BEC. This term determines the stability of the BEC. The particles are repelled from each other when this term is positive. In the opposite case, the particles form clusters. In the case of magnetically ordered systems, they correspond to Suhl or other types of instabilities of homogeneous precession of magnetization. This instability is also observed in other types of magnetically ordered systems. In order to observe BECs in magnets, it is necessary to find a system where the fourth-order term is positive.

Superfluid ^{3}He is a liquid antiferromagnet. Its B phase is a unique three-sublattice antiferromagnet, where term $F_{SO}(|\Psi|^2)$ is zero up to a tilt angle of 104°. At larger angles, it is positive. The BEC appears as soon as the tilt angle becomes larger than 104°, i.e., when the magnon density is high. It is qualitatively different from the atomic BEC. Another phase, ^{3}He-A, has the structure of a two-sublattice antiferromagnet. The term $F_{SO}(|\Psi|^2)$ is negative, and the spatial instability of homogeneous precession is observed [9]. However, it was shown in [10] that when the orbital part of the order parameter is oriented along the magnetic field, this term becomes positive, which makes the BEC possible. It has been discovered in experiments with superfluid ^{3}He-A placed in aerogel (a porous silica structure with 98% porosity) that uniaxial compression of aerogel leads to the orientation of the orbital angular momentum along the compression axis [11, 12]. When the magnetic field was oriented along the compression axis, NMR signals, characteristic of the BEC, were detected [13, 14]. In this case, the magnetization precession frequency depends on tilt angle β as

$$\omega = \omega_L - \frac{\Omega_L^2}{2\omega}\cos\beta, \quad (7)$$

where Ω_L^2 is the square of the frequency of the NMR longitudinal vibration mode.

In the case of small NMR excitation amplitudes in the linear regime, an NMR signal is detected only at the frequency corresponding to the condition $\cos\beta = 1$. When the pumping amplitude is increased, the magnon generation rate becomes larger than the relaxation rate. The equilibrium BEC is formed with a chemical potential corresponding to the pump frequency. In order to satisfy this condition during an increase in the NMR frequency (or a decrease in magnetic field), angle β increases, so that the precession frequency corresponds to the pump frequency. The signal amplitude increases significantly, because the total magnetization precesses coherently at the pump frequency. This BEC state is an eigenstate of the magnetic system. The external pumping determines the chemical potential of the system and, correspondingly, the magnon density. A relatively long-lived NMR signal is observed when NMR pumping is turned off. Its duration is several times longer than the time derived from the inhomogeneity of the external magnetic field [14]. Here, we described BEC formation in the superfluid ^{3}He-A phase, because the free energy and all experimental results of BEC formation in $CsMnF_3$ given in this paper are very similar to those obtained in the ^{3}He-A phase.

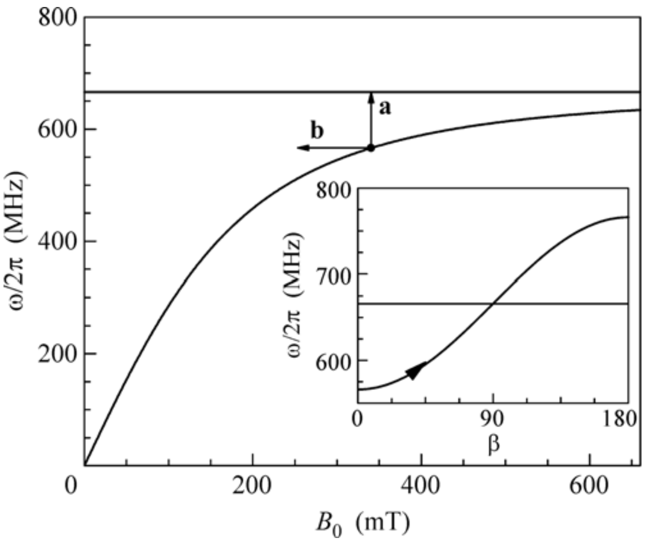

Fig. 1. Frequency of linear ENMR in the $CsMnF_3$ single crystal versus the external magnetic field and (inset) versus the tilt angle of the nuclear magnetization for an external field of 340 mT.

The experiments described below were performed with the $CsMnF_3$ easy-plane antiferromagnet. The magnetic field was directed along the easy magnetization plane. It determined the antiferromagnetic resonance frequency ω_{e0} with the hyperfine interaction Am_eM_n neglected. ^{55}Mn nuclei were in a strong hyperfine field, and the NMR frequency ω_{n0} was on the order of 600 MHz (666 MHz in the low-frequency branch of the NMR in $CsMnF_3$). The hyperfine interaction leads to hybridization of magnetic resonance modes. A hyperfine gap $\Delta\omega_e$ on the order of $7/\sqrt{T}$ GHz emerges in the antiferromagnetic resonance spectrum (where T is the temperature in Kelvin), and this gap depends on the projection of the nuclear magnetization on the electron magnetization axis for each sublattice. The so-called dynamic frequency shift ω_p occurs in the NMR spectrum (pulling). Strictly speaking, both nuclear and electron magnetizations precess in this vibration mode. For this reason, we call it electron–nuclear magnetic resonance (ENMR). The frequency of ENMR ω_{ne} in $CsMnF_3$ in the case of weak excitation and at temperature 1.5 K is shown in Fig. 1. It can be found from the equation

$$\omega_{n0}\omega_{e0} = \omega_{ne}\sqrt{\omega_{e0}^2 + \Delta\omega_e^2}, \quad \omega_p = \omega_{n0} - \omega_{ne}. \qquad (8)$$

In the case of large excitation amplitudes, the frequency of precession depends on the tilt angle of magnetization: $\omega_n(\beta) = \omega_{n0} - \omega_p(M_{nz}/M_n) = \omega_{n0} - \omega_p\cos\beta$, similar to the case of NMR in ^{3}He-A.

The free energy of ENMR under these conditions has the form

$$\mathcal{F} = \int d^3r\left\{\frac{|\nabla\Psi|^2}{2m} + [\omega_{ne}(\mathbf{r}) - \mu]|\Psi|^2 + \frac{1}{2}b|\Psi|^4\right\} \qquad (9)$$

with the chemical potential $\mu = \omega_{rf}$. The fourth-order term is positive and can be written as $b = \omega_p/M_n$.

Correspondingly, the equilibrium density of spin waves for BEC is determined by the difference between the local field and precession frequency:

$$|\Psi|^2 = \frac{\omega_{rf} - \omega_{ne}}{b} = M_n(1 - \cos\beta). \qquad (10)$$

The correct form of the free energy is a necessary, but not a sufficient condition to observe the BEC. First, the relaxation rate of magnons should be sufficiently low, so that they can form the BEC before their relaxation occurs in the case of pulsed NMR. In the case of continuous NMR, magnons should be generated more rapidly than they decay at relatively small pumping amplitudes. The magnetic subsystem should not be overheated. Second, in order to create and maintain a homogeneous BEC state, a sufficient gradient energy is required, which in this case is responsible for spin current emerging in the presence of a wavefunction gradient. This current leads to a redistribution of the magnetization in such a way that all external inhomogeneities are canceled by the inhomogeneity of the local frequency shift, i.e., by a spatial change in angle β. It is important that nuclear–electron precession occurs. The relaxation times of the nuclear subsystem are relatively small. The gradient energy emerges due to the magnetic ordering of the electron spin system. The system under consideration is a very nontrivial magnetic system, in which electron antiferromagnetism is responsible for coherence, and the nuclear subsystem is responsible for the nonlinear term in the free energy.

The emergence of typical BEC signals in $CsMnF_3$ confirms that all these requirements are satisfied. The experiments were performed as follows. The sample was placed in a resonator described in [15] ($Q \sim 200$). Irradiation of the resonator and signal detection were performed using ring antennas, matched with the cavity for 50 Ohm. The main units of the experimental installation were based on the equipment produced by Rohde&Schwarz. We varied the magnetic field, leaving the RF pump power constant. The explanation of BEC formation has been given in the case of changing the RF pump frequency (denoted by arrow **a** in Fig. 1). However, the experiments were performed with changing the magnetic field, like it is usually done for NMR (denoted by arrow **b** in Fig. 1). Figure 2 shows ENMR signals at different RF pump amplitudes. When the excitation amplitude is small, the linear ENMR signal is detected. In the case of large pump amplitudes, when the magnetic field is decreased, the

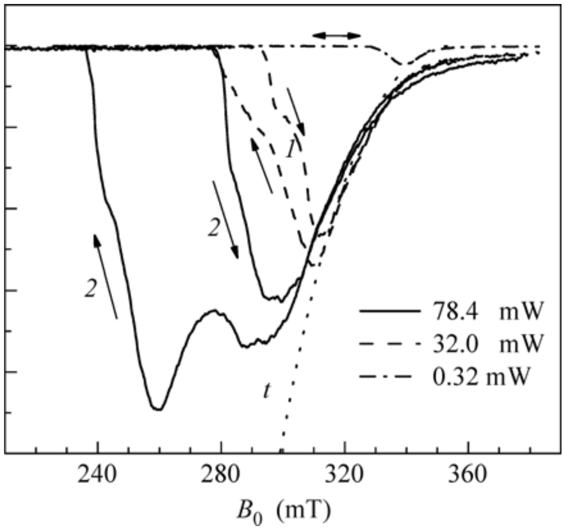

Fig. 2. Signals of ENMR absorption in the $CsMnF_3$ single crystal at different powers of the RF pump field; *t* is the amplitude of the signal, calculated using the condition that the temperature of nuclear subsystem is constant; $\omega/2\pi$ = 566.1 MHz; and T = 1.5 K.

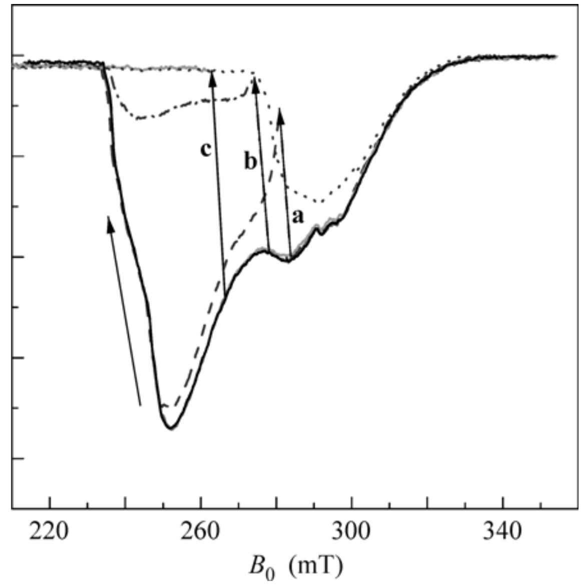

Fig. 3. Signal of BEC evolution in the $CsMnF_3$ single crystal after temporarily switching-off the RF-field; the parameters are $\omega/2\pi$ = 565.4 MHz; T = 1.5 K; and P = 78.4 mW.

precession of the ENMR continues at the RF-field frequency. The magnitude of the signal increases sharply, indicating that BEC forms. Tuning of the ENMR frequency to the RF-pump frequency is performed by changing the number of magnons. When the field is changed in another direction, BEC forms also, but at larger fields. One should keep in mind that the experiments were performed with single crystals, whose magnetic properties are strongly inhomogeneous. There are both domain walls and impurity centers. Similar to the case of ^{3}He-A in aerogel, the BEC is independently present in different parts of the sample. When the tilt angle in a part of the sample is increased, magnons start to decay more rapidly than they are generated or transferred by the superfluid magnetization current. The BEC is destroyed locally. Thus, the net BEC signal decreases. In the backward pass, the BEC signal appears in the fields ensuring the generation of a sufficient number of magnons in this part of the sample. The dynamics of BEC formation at a fixed detuning is very interesting and is shown in Fig. 3. In this experiment, we switched off the RF field for about a second. When the pumping was switched on, the signal returned for thousandths of a second to the value during the backward pass. Then, during a time comparable with that of the total sweep of the field (30 s), the signal returned to its maximum value (Fig. 3a), or BEC formation took place in the part of the sample (Fig. 3b), or BEC did not form at all (Fig. 3c) (depending on the field, when the pumping was switched off). This is evidence that the domain structure of the sample is complex. Such long times of returning to the equilibrium distribution mean that magnons are transferred in the sample at the distances on the order of 1 mm. A more detailed study of the dynamics of BEC formation will be reported later.

We should mention the following result. The BEC state in this case is stable against overheating of the nuclear spin system. The ENMR frequency shift can be reduced both due to a change in the nuclear magnetization and due to the heating of the nuclear spin system. In the latter case, we expected a constant amplitude ENMR signal from the electron system when the field is reduced (similar to the experiments described in [16]). However, in our experiments, we observed an increase in the amplitude of the signal, corresponding to the tilt angle satisfying the condition $\cos\beta = \frac{\omega_{n0} - \omega_{\rm rf}}{\omega_p}$ (curve *t* in Fig. 2). This indicates that the nuclear subsystem is in a dynamic state, and the BEC forms. The experiments on the simultaneous detection of NMR and antiferromagnetic resonance [17] additionally indicate that the temperature of the nuclear subsystem under certain conditions may remain constant.

To conclude, we have detected a new state of magnons in easy-plane antiferromagnet $CsMnF_3$ using the mode of coupled nuclear–electron precession in addition to the earlier known classical BEC states in superfluid ^{3}He [3, 14, 18] and in a ferrite film [19].

We dedicate this work to the memory of Academician A.V.S. Borovik-Romanov, who dreamed of finding magnetic effects in these antiferromagnets similar to those we observed in superfluid ^{3}He. We are grateful to V.A. Panfilov and S.Ya. Khlebnikov for technical assistance. This work was supported by the Russian Ministry of Science and Education, project no. 02.740.11.5217, federal program "Research and Education Staff of Innovative Russia."

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