Magnon Bose-Einstein condensation in CsMnF$_3$ and MnCO$_3$

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Abstract.

The Spin Supercurrent and Bose-Einstein condensation of magnons, similar to an atomic BEC, was observed in 1984 in superfluid $^3$He-B. The same phenomena should exist in solid magnetic systems. We describe here the first observation of magnon BEC in solid easy plane antiferromagnets CsMnF$_3$ and MnCO$_3$. We have observed magnon BEC on a mode of coupled Nuclear-Electron precession. The dynamical properties of this mode have many similarities with NMR of superfluid $^3$He-A. The frequency changes with deflection of nuclear magnetization. Furthermore, the involvement of electron ordered subsystem gives the magnon-magnon interaction, spin waves and spin supercurrent, while the nuclear subsystem gives the relatively long time of relaxation.

The conventional magnon BEC, the phase-coherent precession of magnetization was discovered for the first time in superfluid $^3$He-B in 1984 [1]. It manifests itself by a domain with a fully phase-coherent precession of magnetization, known as the Homogeneously Precessing Domain (HPD). It exhibits all the properties of spin superfluidity: the supercurrent transports of magnetization and phase-slip processes at the critical current [2]; spin current Josephson effect [3]; spin current vortex [4] etc. The comprehensive review one can found in [5] and recent one in [6, 7]. The conventional magnon BEC has been also found in yttrium-iron garnet [8] for the magnons with non zero wave vector.

For a last 25 years there was found 5 different magnon BEC states in superfluid $^3$He [6]. Particularly, the magnon BEC was observed and confirmed in superfluid $^3$He-A immersed in aerogel squeezed along the magnetic field [9, 10]. In this case the aerogel reorients the orbital momentum of $^3$He-A along the field [11, 12]. In this configuration the interaction of magnon is repulsive [13] the coherent precession is stable and the magnon BEC is possible. The same phenomena should exist in solid magnetic systems. Indeed, it was not observed up today. In this article we report the first observation of magnon BEC in solid easy plain antiferromagnets CsMnF$_3$ and MnCO$_3$. The preliminary results were published in [14]. We have observed this phenomena on a mode of coupled Nuclear-Electron precession, which shows the dynamical frequency shift (pulling). Magnetic systems with pulling were studied intensively in 70th of previous century [15]. This phenomenon takes place in the case when the frequencies of NMR and antiferromagnetic resonance are comparable. The typical system with pulling is the easy plane antiferromagnets with ions of Mn. The NMR frequency of Mn$^{55}$ in hyperfine field is very high, about 600 MHz, and the antiferromagnetic resonance frequency in easy
plain and cubic antiferromagnets can be very low. Due to coupling throw the hyperfine field, the two modes of resonance appear, low frequency quasi NMR mode (NEMR) and high frequency quasi antiferromagnetic resonance mode (ENMR). The properties of magnetic subsystems change significantly due to the couple precession. The hyperfine gap appears in a spectrum of antiferromagnetic spin waves. The quasinuclear spin waves appear, with the antiferromagnetic length of coherence. Other words, the nuclear magnetic system get some properties of magnetically ordered system.

The pulse or CW magnetic resonance can move the spin system out of equilibrium. This deviation can be described on the language of elementary excitation, the spin waves (magnons). Magnons can go fast to a new equilibrium state. The local magnetization in this case directed along the magnetic field and transverse components vanished. It means the reheating of the system and the saturation of the signal. Contrary the magnons can remains non-equilibrium for some time. Then they density matrix has a transverse terms. In this case one can see the signal of induction decay, radiates by a precessing spins. The signal can disappear in the case of thermalization or if the spins remains precessing, but its phase became different for the different place of the sample.

Indeed, there is one important question. The nuclear subsystem in CsMnF$_3$ is paramagnetic and after deflection should be dephased due to inhomogeneity of a superfine field. Furthermore, it should follows to Bloch equation. It means, that the nuclear system should thermalize for a time $T_2$ and the transverse component should vanish. In our experiments we have found that it is not a case! In CW NMR experiments the deflected nuclear magnetization precess homogeneously with the constant temperature and the NMR line does not saturated. The explanation can be found in the book [17]. This phenomena was also considered in [18]. There was considered two different scale for inhomogeneity. The microinhomogeneous broadening for a distance smaller then the length of coherence of Suhl-Nakamura interaction (about $10^4$) of the lattice size, and the macroscopic broadening for a longer distances. The microscopic broadening can be suppressed in the case, when pulling is much bigger then a broadening. As a result the antiferromagnetic magnetization precess coherently at a short distance and force the nuclear subsystem precess coherently. For the longer distance it is not the case. It means that the nuclear subsystem shows the properties of ordered one, like the superfluid $^3$He-A. The coherence length is even comparable. The magnetization follows the Landau-Lifshits scenario. It deflects but the module of magnetization does not change.

The density of non-equilibrium magnons depends from the angle of magnetization deflection $\beta$:

$$N = \frac{m - m_z}{\hbar} = \frac{\chi H}{\hbar \gamma} (1 - \cos \beta).$$

(1)

The NMR frequency for $^3$He-A in aerogel dependence on the magnon density (for $\Omega_L << \omega_L$):

$$\Omega = \omega_L - \frac{\Omega_L^2}{2\omega_L} \cos \beta,$$

(2)

where $\omega_L$ and $\Omega_L$ are Larmore and Leggett frequencies and $\beta$ is the angle of magnetization deflection. The frequency dependence for NEMR mode in CsMnF$_3$ and MnCO$_3$ is very similar:

$$\Omega = \omega_n - \omega_p \frac{m_z}{m_0} = \omega_n - \omega_p \cos \beta,$$

(3)

where $\omega_n$ and $\omega_p$ are the non-shifted NMR frequency in hyperfine field and pulling, $m_0$ is the equilibrium nuclear magnetization and $m_z$ its projection on a hyperfine field. It means that the both systems may have a very similar properties and fulfilled the condition of dynamic stability of coherent precession, reads [6, 7]:

$$\frac{\partial \Omega}{\partial \cos \beta} < 0,$$

(4)
Owing this similarity we have suggest to try to observe magnon BEC on a quasi NMR mode in CsMnF$_3$ and CsMnF$_3$[16]

We performed the BEC formation experiments in CsMnF$_3$ and MnC0$_3$ similar to one in $^3$He-A described in Ref.[9, 10]. The frequency shift of Mn$^{55}$ NMR depends from an external magnetic field $H$ in a easy plain of antiferromagnet[18]:

$$\omega_p = \omega_n \frac{H_E H_n m_0}{2H^2 M_0},$$  \hspace{1cm} (5)

where $H_E$ and $H_n$ are exchange and hyperfine fields on an electron sublattice and $m_0$ and $M_0$ are the magnetization of the nuclear and electron sublattices. MnC0$_3$ is a week ferromagnet with the Dzyaloshinskii field about 440 mT. This gives the more complicate, but qualitatively the same equation for pulling.

Figure 1. The schematic presentation of CW NMR spectrometr. The sample is placed inside the resonator. The two loops antennas used for exciting the resonator and to monitor the signal.

Our experiments with CsMnF$_3$ were done at the temperature of 1.5 K at a frequency of 566 MHz. The schema of spectrometer is shown in figure 1. The sample was displaced inside of the resonator. We have used the resonator described in [19]. The one coaxial cable with the loop antenna was used to excite the resonator, and other one to monitor the amplitude of the signal in resonator. The line of CW NMR at small RF excitation corresponds to 437 mT field. If we increase the excitation and sweep down the field, we can see the enormous growing of the signal. The frequency of NMR remains on a frequency of RF excitation even for a magnetic field far away from the resonance field. To explain this phenomena, we should suggest that the frequency shift (pulling) decreases. It can be done by two different way. The temperature may increase or the spins may be deflected on the angle $\beta$ follow the equation:

$$\cos \beta = \frac{\omega_n - \Omega}{\omega_p},$$ \hspace{1cm} (6)

In the first case the NMR signal should saturated. In the second case the signal should grows. Let us suggest that we have a second case and that the all spins precess in phase. Then the amplitude of the signal should follows the equation

$$A = A_0 \sin \beta,$$ \hspace{1cm} (7)
This dependence is shown in figure 2 by dashed line. The amplitude of the signal was calibrated by the amplitude of induction after a short RF pulse. So we can present the signal in the units of angle of deflection $\beta$. There is a fields when all the magnetization precess homogeneously. This is a coherent BEC state. The BEC with deflection angle about $23^0$ have been observed. At further sweep of the field down the RF power can not more compensate the processes of relaxation. The BEC in some part of the sample destroyed and the amplitude of signal decreased. Exactly the same behavior of NMR signal was early observed in superfluid $^3$He-A in aerogel. Also similar behavior we have observed in MnCO$_3$. The results of these experiments are shown on a right side of figure 2. At the sweep back the coherent precession state creates at higher fields.

Finally, we have observed a coherent precession of a magnetization of all the sample. It is not a brute force coherence established by the RF pumping. The amplitude of RF field is only about 5 kHz, while the inhomogeneous broadening of the NMR line at small excitation is about 2 MHz. The external RF field gives the frequency of rotating frame in which the minimum of energy corresponds to a given angle of magnetization deflection (density of magnons). The magnon-magnon interaction and spin supercurrent establishing the coherent precession on a dimensions of the sample. All this phenomena for superfluid $^3$He-A well explained in [9, 10] and reviews [6, 7].

It is important to point out that a similar experiments was described in Ref.[20]. The qualitative behavior of the signal was very similar. Indeed, it is difficult to figure out all parameters of the experiments, described there. To explain those results authors was used the scenario of saturation (heating) of the nuclear subsystem under a RF pumping. This scenario strongly contradicts to our experiments, described here and the other our experiments, which are under preparation for next publication.

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\[ \text{Figure 2. On a left side the amplitude of CW NMR signal from CsMnF}_3 \text{ as a function of external magnetic field for different energy of excitation. The dashed line is a theoretical curve follows the equations (6,7). On the right side shown the similar results measured in MnCO}_3 \]
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