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* In Kazan University the Electron Paramagnetic Resonance (EPR) was discovered by Zavoisky E.K. in 1944.

Double magnetic resonance in $MnCO_3$†

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Results of experiments on $MnCO_3$ investigations by double magnetic resonance are presented. Additional mode of oscillation has been observed in a created Bose-Einstein condensation of magnons state in $MnCO_3$. The properties of observed signals are similar to Goldstone modes.

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1. Introduction

In this article magnetic resonance techniques for the resonance spin system excitation at an additional frequency are presented. This method may be applied when an additional, hidden, mode of resonance exists. The observation of magnetic resonance in Landau field is the best example of this case [1]. The Landau field is Fermi liquid corrections for liquid ^{3}He, incorporated to explain the susceptibility of Fermi liquid. This imaginary field is always directed along the magnetization. Consequently it does not change the Larmor frequency of magnetic resonance. It is just directed along the magnetization, even if the magnetization is deflected and rotates around the external magnetic field. There are two components of magnetization in superfluid ^{3}He - the magnetization of superfluid and normal parts of the liquid. The magnetization of both components are bound to each other and rotates in phase at the temperatures about 0.4 T_c and higher. But at the limit of lower temperatures the two components are unbounded and rotates separately around the common Landau field. The Landau field can be adjusted to the external field by changing the temperature or the pressure. In [1] it was found that the relaxation rate of common Larmor precession increases when the Landau field is equivalent to an external magnetic field. The mode of in-phase precession excites the mode of two components precession around the Landau field. By this experiment it was shown that the Landau field is a real molecular field but not an imaginary.

The double resonance is also observed in the systems with magnon Bose-Einstein condensation (BEC). The magnon BEC is a coherent quantum state of non-equilibrium magnons. It may be created during continuous-wave (CW) nuclear magnetic resonance (NMR) or after a pulsed NMR at the conditions when the minimum of magnon spectrum energy is lower than the chemical potential of excited magnons. In this case the macroscopic number of magnons occupy the lowest energy state according to the Bose statistics. The magnon BEC was first found in superfluid ^{3}He in 1984 [2,3]. The BEC leads to the phenomena of spin superfluidity and related phenomena, like spin current Josephson Effect [4], critical spin supercurrent [5,6],

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spin current Abrikosov vortex [7, 8], etc. All these phenomena show that the BEC state has some rigidity, and consequently the Goldstone and Highs modes of ground state excitation may exist. In the ordered (BEC) state, all spins precess coherently, which means that the whole macroscopic magnetization of the sample of volume V is precessing [9]:

$$M_x + iM_y = M_\perp e^{i\omega t + i\alpha}, \qquad M_\perp = \chi HV \sin\beta \tag{1}$$

where χ is the magnetic susceptibility, α is the phase of precession, β is an angle of magnetization deflection. The spatial oscillation of α corresponds to a new Bosonic excitation, the Goldstone modes of oscillation. These modes have the same nature as in cosmology and the particle physics [10]. The Goldstone modes of oscillation have been found in superfluid ^{3}He-B [11–13].

In this work the BEC state was created by CW NMR. Then the phase modulation h (modulation index) of RF field on a frequency ω_m was applied. When this frequency ω_m is of the order of $\omega_\mathrm{m} = C_{1,2}/2L$, where $C_{1,2}$ is the combination of spin waves velocities at different directions and L is the spatial dimension of BEC state, the additional adsorption is observed. At the moment when ω_m corresponds to one of the Goldstone mode, the BEC signal shows the additional relaxation rate and at sufficient modulation amplitude can even be destroyed. Two different modes of Goldstone oscillation were observed in ^{3}He-B by this method: the axial and plain modes [11–13]. The review of the different experiments with superfluid ^{3}He-B may be found in [9, 14, 15]. There are few different magnon BEC states found in different states of superfluid ^{3}He. One of them is found in superfluid ^{3}He-A [16–18] in the conditions of strong orbital momentum orientation along the magnetic field [19,20]. Exactly the same BEC state was suggested for antiferromagnets with coupled nuclear-electron precession [21]. The BEC state in $MnCO_3$ and $CsMnF_3$ was found in the conditions of CW [22,23], pulsed NMR [24,25] and original switch-off method [26]. The experimental setup for this experiments one can found in [27, 28].

2. Results and discussion

Single crystal $MnCO_3$ was used as a sample in our experiments. The sample was grown by S.V. Petrov in the P.L. Kapitza Institute for physical problems RAS. The crystal has a tablet shape with radius 0.75 mm and 1 mm height. The experiments were performed at the temperature of 1.5 K, at 547.45 MHz frequency and magnetic field of 139.8 mT. Fig. 1 shows the calculated spectrum of nuclear-electron magnetic resonance (NEMR) in $MnCO_3$.

We applied the RF field and swept down the magnetic field. At the point ω_RF the NMR signal appears. In the case of traditional linear NMR the signal should disappear at lower magnetic field values. But in the presence of Suhl–Nacamura interaction the frequency depends on the angle of magnetization deflection [30]:

$$\omega_\mathrm{RF} = \omega_\mathrm{n1} - \omega_\mathrm{p}\cos\beta, \tag{2}$$

where ω_n1 is an unshifted NMR frequency, ω_p is the dynamic frequency shift parameter. This equation has a non-linear solution, when the NMR frequency matches with ω_RF at lower field as $\cos\beta = (\omega_\mathrm{n1} - \omega_\mathrm{RF})/\omega_\mathrm{ne}$. In other words the system may rest at the resonance on the frequency ω_RF even at the lower field if the magnetization deflected on the β angle. This solution is valid only for a region of a sample, where ω_n1 is the same. Usually the spin systems have an inhomogeneous broadening [15]. It means that the local ω_n1 is different for different parts of the sample. Consequently the long standing coherent precession is not possible. The induced precession is still possible but the amplitude of the signal should strongly depends on the amplitude of RF field. Indeed, in our case the amplitude of the signal is extremely large and does not depend on the amplitude of RF field. The signal has critical amplitude of RF field below

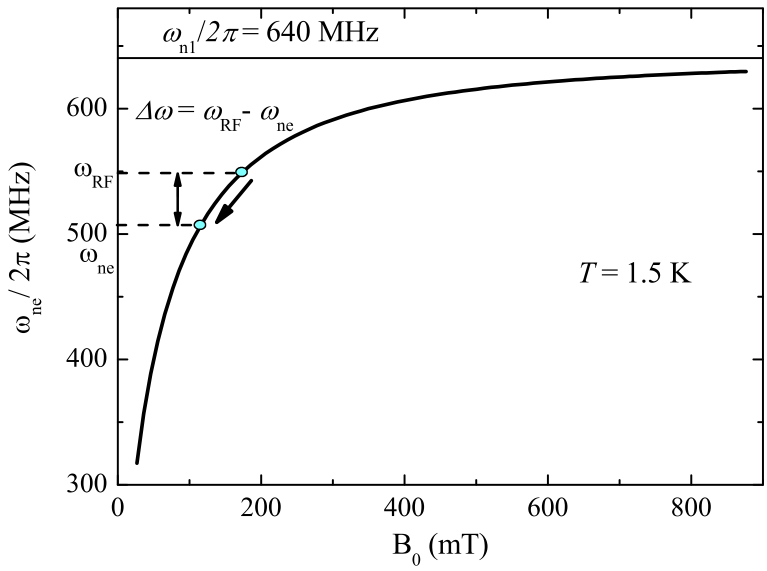

Figure 1. The frequency of NEMR in $MnCO_3$ single crystal as a function of the external magnetic field [29]. The solid horizontal line corresponds to an unshifted NMR frequency. Arrow shows the magnetic field sweep direction in our experiments.

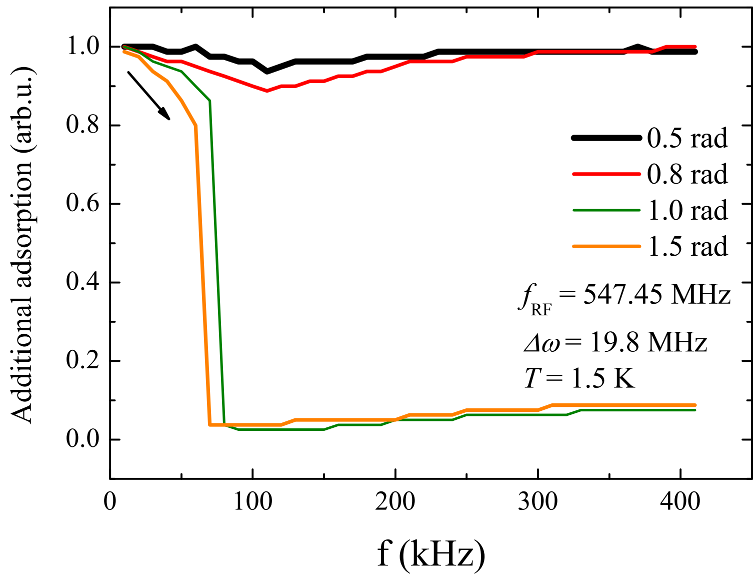

Figure 2. The signal amplitude behavior at different modulation index in $MnCO_3$. Arrows show the direction of phase modulation frequency sweep.

which it disappears. These properties of the signal correspond to a formation of BEC state of magnons in a complete agreement with Bose statistics. If one has pumped the significant number of non-equilibrium magnons with the density $N = S(1 - \cos\beta)$, where $S = \chi H/\hbar\gamma$, the number of magnons becomes bigger than the critical one and the magnons create a single coherent quantum state [9]. The critical angles for magnon BEC was calculated in [31] and correspond to $10°$, which equivalent to $\Delta\omega \approx 2\,\text{MHz}$ for the conditions of our experiments. The next step is to keep the magnon BEC signal at a given field and start to modulate in-phase RF field with a frequency ω_m. In the case of small modulation the small decrease of BEC signal is observed at the frequency about 100 kHz (see Fig. 2). If one increases the depth of modulation, the BEC signal become smaller at this frequency of double resonance. And finally at some critical amplitude of modulation the BEC signal is completely destroyed.

We performed the systematic studies of BEC signal decrease and found that its amplitude decrease linearly with increase of modulation index h, starting from some threshold value of modulation. We may suggest that we excited the Goldstone mode of BEC state. There is not yet clear theory of the Goldstone modes of BEC in $MnCO_3$. Indeed we are able to estimate the frequency of this mode. The velocity of spin waves propagation for a small k is about $C \approx 10^5$ cm/s [32]. The dimensions L of the sample are about 1 mm. The frequency of Goldstone mode is about $\omega_{\mathrm{m}} = C/2L \approx 5 \cdot 10^5$ rad/s ≈ 100 kHz, in the order of the frequency we have observed.

3. Summary

The investigations of single crystal $MnCO_3$ by CW magnetic resonance at the temperature of 1.5 K are presented. The nuclear-electron magnetic resonance signal dependence on the phase modulation index is obtained. The signal properties are very similar to Goldstone modes observed earlier in superfluid ^{3}He-B. The frequency of Goldstone modes is the order of 100 kHz.

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