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

Long-lived free induction decay signal in CsMnF₃ single crystal

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Elementary excitations in antiferromagnets are magnons, and these quasiparticles with integer spin are governed by Bose statistics. In certain conditions their density can be controlled by applied radiofrequency pulse leading to the formation of Bose-Einstein condensation (BEC). We report the investigations of free induction decay signal duration in $CsMnF_3$ under different conditions and discuss it in the framework of the magnon BEC. The observed results in $CsMnF_3$ and previous investigations in superfluid ³He-A are compared.

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1. Introduction

The Bose-Einstein condensation (BEC) corresponds to the formation of a collective quantum state in which macroscopic number of particles is governed by a single wave function. The formation of this state was predicted by Einstein in 1925. The BEC of magnons was discovered experimentally in 1984 in superfluid phase of 3 He-B [1, 2] as a coherent precession of magnetization. Then six different states of superfluid ³He with BEC formation were observed. The review of various experiments on the BEC observation can be found in [3–5]. In all cases BEC is formed by excited non-equilibrium magnons with wave vector k = 0. The pulse or continuous pumping at nuclear magnetic resonance (NMR) frequency was used to excite magnons. In this article we will speak about this canonical case of magnon BEC. The specific of magnon system in solid states makes possible to form a coherent states of magnon with non-zero wave vector. This type of magnon BEC was observed in yttrium iron garnet (YIG) at room temperature by a photon-magnon scattering experiments [6]. In this case non-equilibrium magnons was excited by parametrical pumping on a double frequency. The observation of a similar formation of BEC state of magnons with non-zero k was announced in antiferromagnets in [7] and [8]. In the last case the BEC formation was observed by an induction decay signal, radiated on a double frequency. It was treated as a formation of BEC of pairs of magnons. As follows from our experiments reported in present article, the interpretation of their experiments is not correct. We will describe the correct interpretation of their experiment in the conclusion of this article.

The question of magnon BEC possibility in solid state antiferromagnets was also discussed in [9]. In [10] it was assumed that formation of the BEC state is similar to superfluid ³He, i.e. under direct non-parametrical excitation of quasi-nuclear branch of spectrum, it is possible in solid antiferromagnets $CsMnF_3$ and $MnCO_3$ with coupled nuclear-electron precession. These predictions were successfully confirmed. It was found that the coupled nuclear-electron precession

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shows all properties of coherent spin precession and magnon BEC [11]. The main experimental fact of magnon BEC evidence was independence of the nuclear-electron magnetic resonance (NEMR) signal amplitude on applied radiofrequency (RF) power [12–14]. These experiments were done by means of continuous wave NMR and switch-off NMR. Two regimes of RF pumping were found. In the first regime the induction signal is observed after short (about $1 \mu s$) resonant RF pulse. Let us call this pumped state as a normal. In this state spins precess with its local frequency due to the external magnetic field inhomogeneity. In the second regime the induction signal is observed after long (about hundreds of ms and longer) non-resonant RF pulse (so called switch-off NMR) and the signal amplitude is well described in framework of magnon BEC.

We report here the experimental investigations of the free induction decay (FID) signal behavior in both pumping regimes in two samples of easy-plane antiferromagnet $CsMnF_3$.

2. Results and discussion

Two $CsMnF_3$ single crystals were used as samples in our magnetic resonance experiments. The first one was grown by S.V. Petrov in the P.L. Kapitza Institute for physical problems RAS. The second CsMnF₃ was grown by L.I. Isaenko and S.A. Zhurkov in the V.S. Sobolev Institute of geology and mineralogy SB RAS. Before the expetiment both samples were investigated by X-ray diffraction methods and the first sample is found to have a less homogeneous structure than the second one. The complete details of the experimental setup are available at [15]. All reported experiments were performed at the temperature of 1.5 K. A single RF pulse was applied to the sample and the following FID signal was observed and recorded. Applied RF pulses were homogeneous enough on the scale of the sample to provide confident results. Two pumping regimes were investigated in these experiments. This fact implies that applied RF pulses had different amplitudes, durations and frequencies $f_{\rm RF}$ for various regimes. Short (order of μ) pulses at frequencies within the NEMR line width δf off Larmor frequency f_{Larmor} were used in the normal regime: $\Delta f = f_{\rm RF} - f_{\rm Larmor} \leq \delta f$. And long enough (100 ms - 5 s) RF pulses at Larmor frequencies or higher ones $(\Delta f = f_{\rm RF} - f_{\rm Larmor} > \delta f)$ were used in the second pumping regime, or BEC regime. The typical FID signals in both RF pumping regimes for the first sample are shown in fig. 1a.



Figure 1. The FID signal CsMnF₃ amplitude in the first sample (a) and the second sample (b) in two RF pumping regimes. The amplitudes are normalized to its maximum values. Δf is the frequency shift of RF pumping from the Larmor frequency at given magnetic field. δf is the width of NEMR line measured by continuous wave technique. The increasing in duration of signal in BEC state can be seen.

It can be seen from the plot that the FID signal duration in BEC state after the long 5 s pulse is 2 times longer than for the normal pumped state. Such negligible difference in FID durations between pumping regimes does not allow to analyze the formation of BEC in a proper way for the first sample. The typical FID signals in both RF pumping regimes for the second sample are shown in fig. 1b. It is seen from the plots that in both cases the FID duration describes well by the exponential function. The FID duration time is $0.5 \,\mu$ s in normal state and is about $9.5 \,\mu$ s in magnon BEC state. The FID signal duration in BEC state exceeds one in a normal state for more than an order of magnitude.

In the second (more homogeneous) $CsMnF_3$ sample the signal duration increases more than order of magnitude at transition from one RF pumping regime to another. These results are in full agreement with ones observed in superfluid ³He-A in aerogel [16]. The conclusion about better homogeneity of the second sample is confirmed by FID amplitude investigations [17] where the magnetization was deflected on the bigger angle by comparison with the first sample [14]. The investigation on these samples by continuous wave pumping was presented earlier [11–14] where the independence of the signal amplitude on RF power was observed.

The short FID durations for the first sample can be explained in a following way. It is well known from the experiments with ³He that process of magnon BEC state formation is very sensitive to impurities and magnetic defects. For example, the formation of a BEC state of ³He-B in aerogel (high porosity SiO₂) was observed for the first time in Grenoble as a formation of a signal localized in some local region of the sample [18]. Later a global BEC signal for the sample with a better homogeneity was observed [19]. The same phenomena was observed in a pulsed NMR [20] as well. The FID of pure ³He-B may have duration of a seconds, while from the ³He-B in aerogel it lasts only few milliseconds. The similar behavior of the FID signal was observed in superfluid ³He-A in aerogel. The FID duration in BEC state was only about an order of magnitude longer than in the normal state. Furthermore, the beating of a FID signals from a few local BEC states was observed in short pulse NMR. In the case of switch-off NMR the beats is not observed and the duration of the signal in BEC state increases [16]. From another point of view, it is known [21] that the relaxation times of CsMnF₃ crystals can vary more than order of magnitude. Thus the short FID durations of the first sample supposed to come from impurities or defects of the single crystal.

3. Summary

We performed experiments on formation of magnon BEC in CsMnF₃ antiferromagnets. This classical BEC radiates the resonance induction decay signal, as well as a double frequency signal. The formation of coherent magnon pairs does not follow from the theory so it cannot be applied for explaining the experimental results described here. We have investigated the duration of the FID signal in two samples of easy-plane antiferromagnet CsMnF₃. In addition to our previous investigations of the signal amplitude by means of continuous wave NMR and pulse NMR these results also confirm the formation of magnon BEC in antiferromagnet CsMnF₃. In more homogeneous sample in coherent precession state we see the long-lived induction decay signal which duration is longer than in the first low quality sample. Thus the search of homogeneous samples of good quality is a key task for exact detection all the properties of magnon Bose-Einstein condensation in solid antiferromagnets. Our experiment shed light on the results of spin waves showed here, the excitation of spin waves leads to a spin waves spectrum shift. As a result the spin waves with k = 0 appear and form the conventional BEC.

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