

# Magnon BEC in Antiferromagnets with Suhl–Nakamura Interaction

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**Abstract** From atomic physics one knows the phenomenon of Bose–Einstein condensation (BEC), where a macroscopic ensemble of particles occupy coherently a single state. Similar phenomena were observed for different types of quasiparticles in condensed matter. Here we present the results of investigations on the BEC of elementary magnetic excitations—magnons—in antiferromagnets with a dynamical frequency shift.

**Keywords** BEC of quasi-particles · Nonlinear NMR · Spin superfluidity

## 1 Introduction

Predicted in 1925 by Einstein for bosonic particles (see e.g. [1]), the phenomenon of Bose–Einstein condensation (BEC) corresponds to the formation of a collective quantum state where a macroscopic number of particles is governed by a single wave function. Almost perfect BEC states were observed in ultra-cold atomic gases. In Bose

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liquids, BEC is strongly modified by interactions, but still remains the key mechanism for the formation of a coherent quantum state which experiences the phenomenon of superfluidity, i.e., the existence of a non-dissipative superfluid current. Both BEC and superfluidity do not require a strict conservation of the number of particles. These phenomena can also be observed even if such a conservation law is weakly violated (for example in systems of such sufficiently long-lived quasiparticles as phonons, rotons, spin waves (magnons), excitons, etc.), when the life time of a quasiparticle is significantly longer than its thermalization time.

The phenomenon of magnon BEC has been described by Bunkov and Volovik [2, 3] in order to explain long-lived induction decay signals and unusual spin dynamics in superfluid  $^3\text{He}$ . Magnon BEC is a direct magnetic analogue of atomic BEC. It is manifested by the phase-coherent precession of magnetization (even in an inhomogeneous static magnetic field) which was discovered experimentally in superfluid  $^3\text{He-B}$  [4]. The first theoretical explanation of spin superfluidity was given in Ref. [5]. In the BEC state, the transverse component of magnetization is described by a wave function  $S_{\perp} \exp(i\omega t + \phi)$  which demonstrates all the properties of spin superfluidity. The spatial gradient of the phase  $\phi$  leads to a spin supercurrent transporting the magnetization. The experimental observations include phase-slip processes at a critical current [6], the Josephson spin-current effect [7], as well as spin-current vortices [8], Goldstone modes [9–11] etc.

During the last 25 years, five different magnon BEC states were found in the  $^3\text{He}$  superfluids. Comprehensive reviews can be found in Refs. [12–14], while more recent accounts are Refs. [15–17]. For the purpose of this report, the BEC state in superfluid  $^3\text{He-A}$  is the most interesting one. Homogeneous precession of the magnetization in bulk  $^3\text{He-A}$  is not stable due to the attractive interaction between the magnons. Such an instability was predicted theoretically [18] and also observed experimentally [19, 20]. Later it was suggested by Bunkov and Volovik [21] that a reorientation of the orbital momentum of  $^3\text{He-A}$  could lead to a repulsive interaction between the magnons and to their BEC formation. This suggestion was confirmed in experiments with  $^3\text{He-A}$  immersed in aerogel which had been squeezed along the magnetic field. In this case, aerogel orients the orbital momentum of  $^3\text{He-A}$  along the magnetic field [22, 23]. In such a configuration the magnon BEC was observed [24, 25]. This observation is important for the subject of this paper because the dynamic properties of NMR in antiferromagnets (AFMs), as discussed here, are similar to the dynamic magnetic properties of superfluid  $^3\text{He-A}$  in squeezed aerogel. Moreover, the magnetic interaction energy is similar to that responsible for BEC in cold atom clouds [26].

From the magnetism point of view, superfluid  $^3\text{He}$  exists in a peculiar antiferromagnetic state. Its spin superfluidity is a consequence of the particular magnetic interaction and is not directly related to mass superfluidity. This is the reason why the search for new classes of magnetic materials showing the same type of phenomena is an important project [27]. The magnon BEC, similar to that in  $^3\text{He-A}$ , was found in the easy-plane AFMs  $\text{CsMnF}_3$  and  $\text{MnCO}_3$  with a strong electron-nuclear interaction [28, 29]. BEC of magnons was also found to apply for parametrically excited magnons in yttrium-iron garnet [30] and in AFMs [31] for magnons with non-zero wave vector. In this report we describe investigations of magnon BEC with  $k = 0$  in  $\text{CsMnF}_3$  and  $\text{MnCO}_3$  antiferromagnets.

## 2 Coupled Nuclear-Electron Precession

The term nuclear spin wave (or nuclear magnon) applies to elementary excitations in coupled electron and nuclear spin systems in the nuclear magnetic resonance (NMR) frequency range. The most remarkable property of these excitations is that they represent coupled oscillations of two completely different magnetic subsystems. Electron spins are ordered by an exchange interaction while the nuclear spins are in the paramagnetic state. As a result of combined oscillations of these two subsystems, the frequency of the electron magnons increases, while the NMR frequency decreases and becomes significantly lower than the Larmor frequency of the nuclear spins. In a good approximation, the non-shifted frequencies of the nuclear and electron resonances  $\omega^{n0}$  and  $\omega^{e0}$  and the shifted frequencies  $\omega^n$  and  $\omega^e$  can be described by the following relation

$$\omega^{n0} \omega^{e0} = \omega^n \omega^e. \tag{1}$$

The energy of interaction between the nuclear and electron branches is determined by the hyperfine field resulting from the nuclei acting on the electrons

$$H_{hf}^e = A \gamma_e m_z = A \gamma_e m_0 \cos \beta, \tag{2}$$

where  $m_z$  is the projection of the nuclear magnetization onto the electron magnetization, which is controlled by the nuclear magnetization  $m_0$  and may be reduced by heating or by deflecting the angle  $\beta$ . In the latter case, we may speak about an interesting non-linear phenomenon, i.e., the frequency dependence of the spin excitation, as was theoretically discussed by de Gennes et al. [32,33]

$$\omega^n = \omega^{n0} - \omega_{p0} \cos \beta. \tag{3}$$

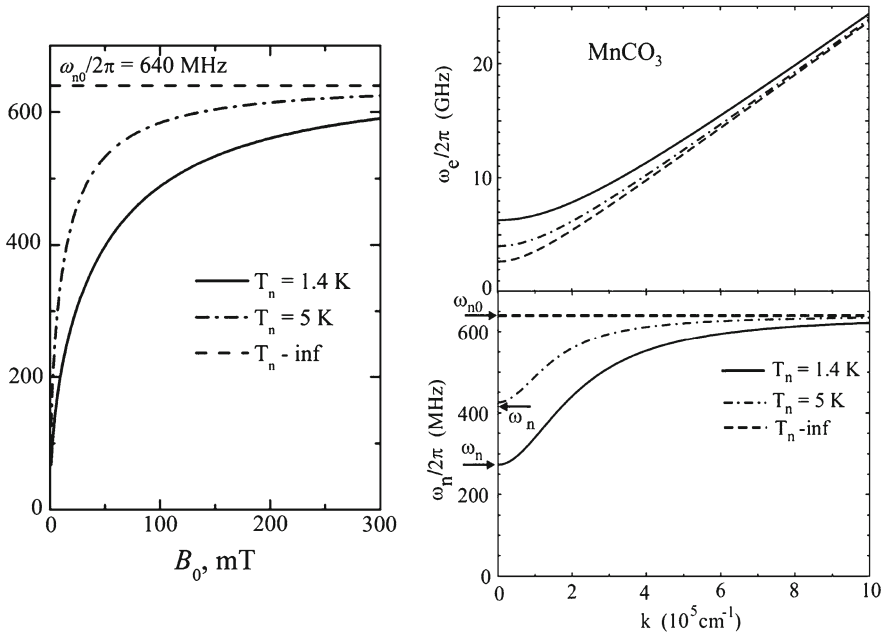
This dynamical frequency shift of the NMR, so-called “pulling” possesses many similar properties with the frequency shift in superfluid  $^3\text{He-A}$ : The NMR frequency depends strongly on the deflection angle of the magnetization. The spin systems considered in this paper can be described in terms of the Suhl–Nakamura interaction [34,35]. It is an indirect nuclear-nuclear exchange interaction via the magnetically ordered electrons. For the  $\text{CsMnF}_3$  and  $\text{MnCO}_3$  AFMs the NMR frequency  $\omega^{n0}$  of  $^{55}\text{Mn}$  is very high, being about 600 MHz, while the electron *AFM resonance* frequency  $\omega^{e0}$  might be very low at small external magnetic fields  $H$ . The set of equations for these AFMs can be written in a good approximation as follows [36]

$$\omega_k^e = \sqrt{(\omega^{e0})^2 + (\omega_{hf}^e)^2 + k^2 v_s^2}, \quad \omega^{e0} = \gamma_e \sqrt{H(H + H_D)},$$

$$\omega_{hf}^e = \gamma_e \sqrt{2H_E H_{hf}^e}, \quad H_{hf}^e = A \gamma_e m_z, \tag{4}$$

$$\omega_k^n = \omega^{n0} - \frac{\omega_p}{1 + (kr_0)^2}, \quad \omega^{n0} = \gamma_n (H + H_{hf}^n),$$

$$\omega_p = \omega^{n0} \frac{H_E H_{hf}^e}{H(H + H_D)} \frac{m_0}{M_0}, \quad H_{hf}^n = A \gamma_n M. \tag{5}$$



**Fig. 1** *Left* The frequency of NMR in  $\text{MnCO}_3$  at low resonance excitation at the temperatures of 5 and 1.4 K as a function of the external magnetic field for  $k = 0$ . *Right* The spectrum of spin waves at 20 mT field at the same temperatures

Here  $\gamma_e$  and  $\gamma_n$  are the gyromagnetic ratios of the electrons and of the  $^{55}\text{Mn}$  nuclei, respectively,  $H$  is the external magnetic field and  $H_D$  is the so-called Dzyaloshinsky magnetic field ( $H_D \simeq 4.4$  kOe in  $\text{MnCO}_3$  and zero in  $\text{CsMnF}_3$ ),  $H_E$  is the exchange field,  $H_{hf}^e$  and  $H_{hf}^n$  are the hyperfine magnetic fields acting on the electron and nuclear sublattices, respectively, while  $A$  is the hyperfine constant,  $M_0 \equiv |M|$  and  $m_0 \equiv |m|$  are the electron and the nuclear magnetizations, respectively. Finally,  $r_0$  stands for the Suhl–Nakamura interaction radius (about  $10^4$  interatomic distances for the AFMs considered here) which originates from the stiffness of the magnetically ordered electron system. The magnetic field dependence of the frequency of the quasinuclear branch  $\omega_k^n$  with  $k = 0$  is shown in Fig. 1 on the left, while the magnon spectrum of both branches  $\omega_k^e$  and  $\omega_k^n$  shown in Fig. 1 on the right.

Owing to the non-linearity of the frequency dependence, the magnetization cannot be deflected by a large angle, since its frequency turns off from the resonance condition during the RF pulse. In Refs. [32,33] it was argued that the usual spin echo signal is not formed in this case. However, this argument would be valid only for the classical Hahn mechanism of echo formation. For systems with pulling another mechanism of echo formation was suggested [37], namely the frequency modulation echo. It takes place due to frequency modulation from the inhomogeneous broadening of the spin precession after applying the second pulse. This mechanism of echo formation, as well as that for a single pulse echo, [38] confirms that the NMR frequency depends on the angle of the nuclear magnetization deflection. Moreover, the frequency shift as a

function of the magnetization deflection was observed directly in experiments where another type of echo formation (called Bunkov echo [39]) was taking place. This kind of echo is observed after two pulses. The first pulse is applied in resonance conditions, while the second obeys parametrical excitation [40,41].

The magnetic system obeying Eqs. (4–5) is very unusual. There is a common precession of both the magnetically ordered electron spin and the paramagnetic nuclear spin systems. This means that the electron spins, when deflected by the RF pulse, cannot dephase over the distance of the Suhl–Nakamura interaction radius  $r_0$ . The deflected electron spins induce an RF field on the nuclei  $H_{hf}^n = A\gamma_n M \sin \alpha$ , where  $\alpha$  is the deflection angle of the electron magnetization. The local hyperfine field acting on the different nuclei may vary by  $\Delta H_{hf}^n$  due to localized impurities. In the case when the pulling frequency  $\omega_p$  is comparable with  $\gamma_n \Delta H_{hf}^n$ , the nuclear spins at a distance larger than  $r_0$  may dephase from the common nuclear precession. Even a small fraction of these spins may lead to fast relaxation  $T_2$  of the precessing magnetization. To escape this mechanism of relaxation, the condition  $\omega_p \gg \gamma_n \Delta H_{hf}^n$  should be fulfilled [42,43]. It was found experimentally that our nuclear spin system is coherent when  $\omega_p > 30$  MHz.

As mentioned above, the nuclear spin system in easy-plane AFMs has many similarities with superfluid  $^3\text{He-A}$  [26]. In both cases, the frequency of long-wave-length magnons includes positive quadratic contributions  $\delta\omega_k \propto k^2$ . Also an increase of the nonlinear frequency shift is observed as a function of the excitation level (due to a repulsive interaction between magnons). Taking into account that the magnon BEC in  $^3\text{He-A}$  is observed under continuous wave NMR conditions, [24,25] we carried out similar experiments in  $\text{CsMnF}_3$  and  $\text{MnCO}_3$  and indeed observed very similar results [28]. In particular, we observed the formation of a NMR signal of huge amplitude while the magnetic field was swept downward. This signal appeared in the conditions where  $\omega_{RF} = \omega^{n0} - \omega_{p0}$ . During further downward sweeping the NMR signal amplitude, i.e. the module of the transverse nuclear magnetization  $I = \sqrt{I_x^2 + I_y^2}$ , grew proportional to  $\sin \beta$ , where

$$\beta = \arccos \frac{\omega^{n0} - \omega_{RF}}{\omega_{p0}(H)}. \tag{6}$$

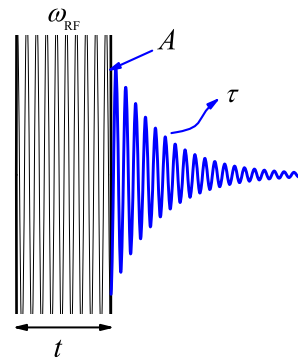
This means that the nuclear spins precess at the frequency of the RF field. During a downward field sweep  $\omega_{p0}(H)$  grows, but the NMR frequency does not change due to the deflection of the nuclear magnetization by the angle  $\beta$ . This condition corresponds exactly to the energy minimum of the spin system in the rotating frame, as was shown in Refs. [44,45].

Here we present new experimental results on the study of magnon BEC in  $\text{CsMnF}_3$ , observed on a new (and better quality) crystalline sample of  $\text{CsMnF}_3$ .

### 3 Experimental Results

It is obvious that the main requirement is the high quality of the investigated samples. For the formation of the magnon condensate state we need a sufficiently long

**Fig. 2** The scheme of the experiment. The *black line* shows the RF pulse applied to the sample. The *blue line* shows the free induction decay signal (Color figure online)

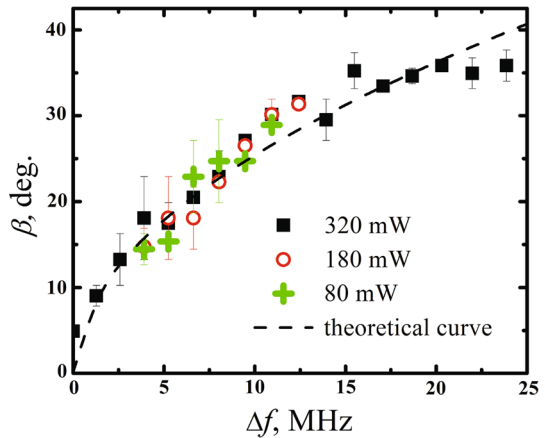


spin-lattice relaxation time as well as the absence of signal beating between different parts of the sample. The present sample of  $\text{CsMnF}_3$  was prepared in the V.S. Sobolev Institute for Geology and Mineralogy (of the SB of RAS) by L.I. Isaenko and S.A. Zhurkov in 2012. X-ray analysis shows that this new sample is more homogeneous than our previous sample of  $\text{CsMnF}_3$  in which we earlier observed the formation of the magnon BEC state [29]. Our experiments were done at a temperature of 1.5 K. The sample was placed inside a resonator tuned to the frequency  $\omega_{\text{RF}} = 562.55$  MHz with a  $Q = 150$ . Complete details of the experimental setup were published in Ref. [46]. A simplified scheme of the experiment is presented in Fig. 2. A high power RF pulse is applied to the sample and the free induction decay (FID) signal is monitored. The main feature of our setup is that one may vary the duration of the RF pulse over a wide range (from hundreds of nanoseconds to a few seconds). Thus we were able to perform both pulsed NMR experiments (RF pulse duration is less than the relaxation time) and switch-off NMR experiments (RF pulse duration is longer than the relaxation time) with the same experimental setup.

As known from experiments with  $^3\text{He-A}$ , the process of magnon BEC formation is very sensitive to impurities and magnetic defects. For example, the formation of a BEC state in  $^3\text{He-B}$  in aerogel was observed for the first time in Grenoble as a formation of a signal localized in a certain region of the sample [47]. A global BEC signal was later observed in a sample of better homogeneity [48].

To enhance the conditions for BEC in  $\text{CsMnF}_3$ , we use a new sample prepared in the V.S. Sobolev Institute for Geology and Mineralogy of SB RAS. We observed the induction decay signal not only after short pulses ( $t \leq T_1, T_2$ ) but also after long pulses ( $t > T_1, T_2$ ). In certain conditions, the signal amplitude after a long pulse was even larger than that after a short resonance pulse [29]. The large-amplitude signal was observed when the frequency of RF pumping was significantly higher than the resonance frequency ( $\omega_{\text{RF}} > \omega_k^n(0)$ ). After a short RF pumping we see the usual induction signal only in resonant conditions ( $\omega_{\text{RF}} \approx \omega_k^n(0)$ ). When the RF pulse duration is increased, a transition process occurs in the sample after which the FID amplitude reaches an equilibrium value and remains unchanged during further increase of the RF pulse duration. The same FID signal is also observed far from the resonant conditions, but, in this case, the RF pumping needs to be of sufficiently long duration. In such cases, definitely, a stationary state of magnetization (in the rotating frame) is

**Fig. 3** The free induction decay amplitude (in units of the magnetization deflection angle  $\beta$ ) in the new sample of CsMnF<sub>3</sub> after a 10 s pulse for three different RF power levels as a function of frequency shift at a temperature of 1.5 K. The dashed black line shows the theoretical curve corresponding to the minimum of the magnetization energy in the rotating frame, where the BEC is formed (Color figure online)



formed. This state corresponds to the minimum of magnetic and spectroscopic energy, as explained in Ref. [29].

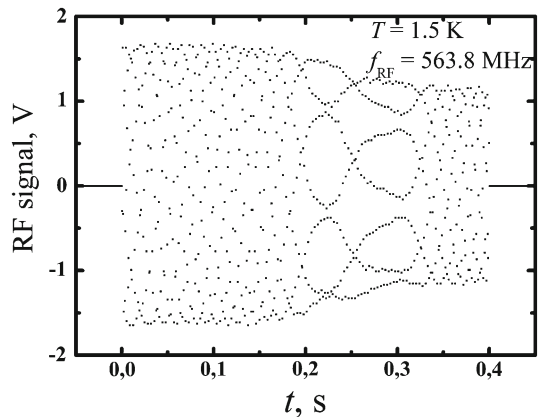
In Fig. 3 the FID amplitude is shown (in units of the deflection angle of the magnetization) for the new better quality sample after a long RF pulse, as a function of the frequency shift at different power levels of RF pumping. One can see that the signal amplitude does not depend on the RF pumping power but is completely defined by the frequency shift. This can be identically rewritten as a condition for the formation of BEC of magnons:

$$\omega_{\text{RF}} = \omega_0^n(\beta) = \omega^{n0} - \omega_p(0) \cos \beta . \tag{7}$$

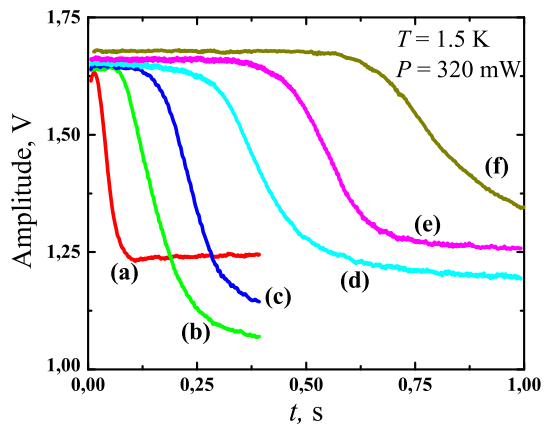
We have found that condition (7) corresponds exactly to our experimental results. We have here the possibility to deflect the magnetization by larger angles than with our earlier sample. In particular, the deflection angle was greater than 30° at a frequency shift of 24 MHz. The maximal deflection angle in previous experiments with the old sample was about 21° at a frequency shift of 9 MHz [29]. In this work we investigated the new high-quality sample and were now able to pump more magnons. This shows that the quality of the sample is the key factor for a clear observation of the magnon condensate state. The larger initial frequency difference increases the FID signal whose amplitude is in good correspondence with the angle  $\beta$  for the conditions of BEC formation at a given frequency and magnetic field.

Let us consider what happens with the magnetic system of CsMnF<sub>3</sub> during the application of a long non-resonant RF pulse. The details of the experimental setup for these investigations were discussed in Ref. [46]. We apply a long non-resonant RF pulse to the transmitter loop antenna, which excites the resonator with the CsMnF<sub>3</sub> sample. Part of the RF pulse power is absorbed by the sample and we measure the decrement of the RF amplitude in the detector loop antenna. An example of the RF signal in the detector antenna is shown in Fig. 4. The time moments of  $t=0$  and  $t=0.4$  s show the beginning and the end of the RF pulse applied to the transmitter antenna. One can see that the transition process, manifested by the decreasing RF amplitude,

**Fig. 4** The digital record of the signal seen in the detector antenna during an RF pulse. The moment  $t=0$  is the start of the RF pulse applied to the transmitter antenna. The moment  $t=0.4$  s corresponds to the end of the applied RF pulse. Every point corresponds to about 500 oscillations



**Fig. 5** The amplitude of the RF signal in the detector loop antenna at different frequency shifts. Curve *a* corresponds to the frequency shift 27 MHz, *b* –33 MHz, *c* –35 MHz, *d* –40 MHz, *e* –42 MHz, *f* –45 MHz (Color figure online)



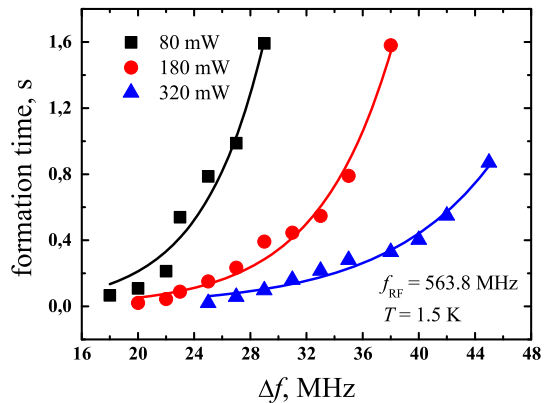
occurs on a time scale of about 250 ms at the frequency shift  $\Delta\omega = \omega_{\text{RF}} - \omega_k^n(0) = 35$  MHz. This time is reached approximately in the middle of the RF pulse in Fig. 4.

This time scale is much longer than any relaxation process in the system. At the same time, this time scale is comparable to the RF pulse durations after which the magnon BEC radiation is observed. That is why the transition process shown in Fig. 4 can be explained as magnon condensate formation.

It can be seen from Fig. 5 and Fig. 6 that the time for the formation of the magnon condensate depends on both the RF power and the frequency shift. It turned out that the formation time of the magnon BEC state during RF pumping increases with the frequency shift. The observed phenomena of BEC formation have a straightforward explanation. At the beginning of RF pumping spin waves are excited with their wave vector  $k$  corresponding to the condition  $\omega_{\text{RF}} = \omega_k^n$ . The direct process is possible due to the inhomogeneities in the sample over a distance  $1/k$ . The number of magnons grows exponentially. For magnons, there are two possibilities. They might thermalize which means that they occupy the whole spectrum of spin waves. They may also condense in the ground state with  $k = 0$ . If the former process takes place, one would not be able to see the induction signal after switching off the RF field. In the latter case



**Fig. 6** The time of magnon condensate formation during the RF pulse, shown as a function of the frequency shift at various values of pumping power (Color figure online)



transverse magnetization appears. The number of the ground-state magnons corresponds to  $n = m_0(1 - \cos \beta_m)$ . The frequency of the precessing magnetization in the ground state changes as given in Eq. (3). This process will stop when  $\beta_m$  fulfills the condition  $\omega_{\text{RF}} = \omega_0^n(\beta) = \omega_0^n - \omega_p(0) \cos \beta_m$ . This means that the chemical potential of magnons corresponds to the chemical potential of the magnetization in the rotating frame. In this case, an induction decay signal with the amplitude  $m_0 \sin \beta$  appears when the RF pumping is switched off. This signal we have observed.

## 4 Conclusions

The conditions of magnon BEC depend strongly on magnon relaxation. In our new sample of CsMnF<sub>3</sub> the rate of relaxation is smaller by a factor of 2 to 3. As a result, we were able to investigate directly both magnon pumping at  $k \neq 0$  and condensate formation in the ground state with  $k = 0$ . These processes explain the formation of the measured induction decay signal after a long RF pulse, which is much longer than the magnetic relaxation.

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