Double acoustic nuclear antiferromagnetic resonance in $RbMnF_3$ and $KMnF_3$

Kh. G. Bogdanova, V. A. Golenishchev-Kutuzov, F. S. Vagapova, A. A. Monakhov, and R. V. Saburova

Kazan' Physico-technical Institute, USSR Academy of Sciences
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Results are given of experiments carried out by a new method of studying acoustic nuclear magnetic resonance in magnetic crystals—by double acoustic nuclear antiferromagnetic resonance. Absorption of acoustic energy by the spin system of Mn^{55} nuclei in $RbMnF_3$ and $KMnF_3$ is detected by the field shift of the antiferromagnetic resonance. The effective nuclear spin system temperature is determined as a function of the sound frequency and of the magnetoelastic coupling constant.

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INTRODUCTION

It is known that mixing of the vibrations of electron and nuclear spins takes place in ferro- and antiferromagnets due to hyperfine interaction.[1-6] The dynamic coupling of the characteristic resonance frequencies of the electron and nuclear spin systems has been thoroughly studied in a number of cubic antiferromagnets ($RbMnF_3$, $MnCO_3$, $KMnF_3$, $CsMnF_3$), in which this coupling is especially strong. Witt and Portis[7] and Ince[8] have observed a number of interesting nonlinear phenomena in these crystals by the method of double electron-nuclear resonance. Absorption of energy by the nuclear spins, recorded by the shift in the resonant magnetic field H_0 for the antiferromagnetic resonance (AFMR) was observed in a range of frequencies that greatly exceeds the width of the nuclear magnetic resonance (NMR) line; a threshold of pump power for the AFMR shift and a dependence of the shift on the power as the nuclear frequency approaches the hyperfine frequency were both observed.

The present paper reports on the first experimental investigation of the double acoustic resonance in antiferromagnets that was predicted by Turov and Petrov.[6] It is well known that the methods of ultrasonic spectroscopy are especially effective in the investigation of spin-phonon interactions in magnetic materials. In ferro- and antiferromagnets, the method of acoustic NMR or the double resonance method used by us can be employed for effective study of the magnetoelastic coupling of electron-nuclear interactions and the coupling of the spin systems with the lattice. The ultrasonic methods allow us to distinguish the mechanisms of interaction, inasmuch as the latter depend on the direction of propagation and the polarization of the sound, and this creates additional advantages over electromagnetic excitation. Moreover, in contrast with the usual NMR, the acoustic methods mentioned above allow us to study both branches of the nuclear spin oscillations[9], since the coefficients of amplification for both modes are of the same order ($\sim 10^2$).

METHOD AND RESULTS OF MEASUREMENT

Experiments on the double acousto-magnetic resonances were carried out at 4.2°K on single crystals of $RbMnF_3$ and $KMnF_3$ prepared in the shape of parallelepipeds with dimensions $4 \times 6 \times 5$ mm ($RbMnF_3$) and $4 \times 6 \times 4$ mm ($KMnF_3$), the sides of which were disposed along the crystallographic axes, while the faces were mutually parallel to ~(20–25) sec. Both materials have a cubic structure. Below the Neel point, $RbMnF_3$ ($T_n = 82.6$°K) goes over into the antiferromagnetic state, while $KMnF_3$ exhibits weak antiferromagnetism below $T_n = 88.3$°K. Their basic feature is the combination of small anisotropy fields ($H_A(RbMnF_3) = 3.8$ Oe, $H_A(KMnF_3) = 2.9$ Oe) and large hyperfine fields ($H_N(RbMnF_3) = 6.52 \times 10^5$ Oe, $H_N(KMnF_3) = 6.5 \times 10^5$ Oe).

The experiments were carried out on an apparatus based on a standard EPR-3 radiospectrometer (Fig. 1). The samples 1 were placed on the lower wall of a TE_{101} rectangular resonator, made from a length of standard silver-plated three-centimeter waveguide-band.

An acoustic head was mounted on the lower end of the sample. The propagation vector of the sound wave **k** and the constant magnetic field H_0 were orthogonal to each other.

We used surface excitation to excite acoustic oscillations at the frequencies of the nuclear transitions of Mn^{55} (500–700 MHz). Single crystals of lithium niobate 3 were used as the piezoelectric transducers; these crystals are piezoferroelectric with a very large electromechanical coupling coefficient. The piezotransducers were made in the shape of parallelepipeds with dimensions $6 \times 4 \times 12$ mm. All the surfaces were polished to be plane and parallel to within a few seconds.

With the help of special electrodes, attached to the surface and connected to the high-frequency electromagnetic generator, either longitudinal or transverse waves

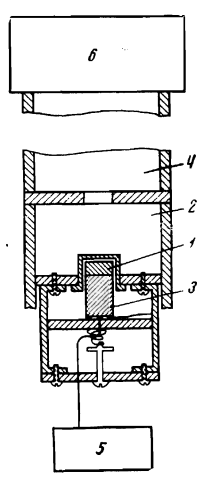

FIG. 1. Block diagram of spectrometer for acoustic nuclear antiferromagnetic resonance. 1–sample, 2–resonator, 3–piezoelectric transducer, 4–waveguide, 5–pump generator, 6–AFMR spectrometer.

were excited in the lithium niobate single crystal. The acoustic oscillations were transferred to the sample of $RbMnF_3$ or $KMnF_3$ under study through a thin layer of metallic indium or special low-temperature grease. In order not to disrupt the parallelism between the surfaces of the sample and the transducers, uniformity of the bond thickness was maintained accurate to 20–25 sec. The working resonator with the sample and the acoustic head was placed in a metallic helium cryostat. The sample was immersed in the liquid helium in order to lower the radiofrequency heating. Special measures were taken to shield the sample from the effects of the electromagnetic waves exciting the piezotransducer. Trial experiments showed that the AFMR shift is accomplished by the acoustic and not the electromagnetic field.

The method of investigation consisted of the following. The antiferromagnetic resonance signals at the frequency ν_A = 9140 MHz were observed by varying H_0. The resonance values of the fields were $H_0(RbMnF_3)$ = 3546 Oe, $H_0(KMnF_3)$ = 2600 Oe for the (+) mode and $H_0(RbMnF_3)$ = 1049 Oe and $H_0(KMnF_3)$ = 1544 Oe for the low-field (−) mode. The field of the nuclear pump was then switched on. Acoustic saturation was produced in the 540–700 MHz range and the AFMR signals were then observed.

In both compounds, a shift was observed in the resonance field H_0 for the field-dependent antiferromagnetic mode (ω_{e+}) on excitation of longitudinal acoustic waves in the samples with a power of 10^{-3} W/cm^2. The H_0 shift for $RbMnF_3$ takes place in the range of nuclear-pump frequencies from 600–685 MHz, and for $KMnF_3$ in the range 590–675 MHz.

The upper part of Fig. 2a shows the first derivatives of the AFMR lines in $RbMnF_3$ without pumping for the (−) and (+) modes, and the lower part of the same lines

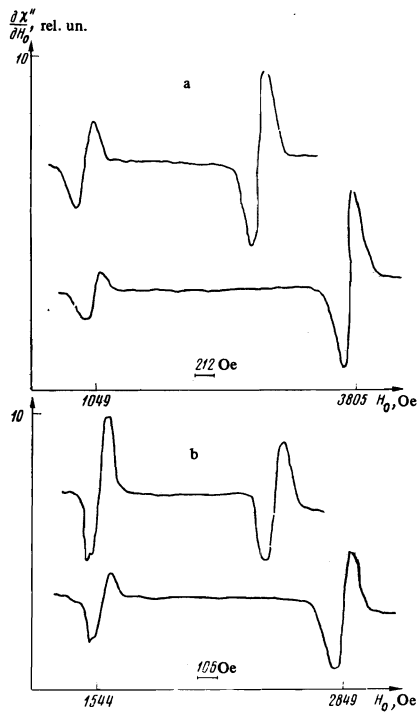

FIG. 2. First derivative of the AFMR signals without acoustic pump (above) and with acoustic pump (below): a–for $RbMnF_3$, b–for $KMnF_3$;

for acoustic pumping. Figure 2b shows the same curves for $KMnF_3$ ($H_0^{(+)}$(without pumping) = 2600 Oe and $H_0^{(+)}$ (with sound) = 2849 Oe).

At a constant sound power and increasing pump frequency, an increase in the AFMR shift is observed first for the (+) mode (up to about 640 MHz) and then a decrease (Fig. 3a for $RbMnF_3$ and Fig. 3b for $KMnF_3$). However, if the sound power is increased, the shift increases steadily up to the hyperfine frequency ω_N.

A decrease by almost a factor of two was also observed in the intensity of the low-field (−) mode of the AFMR (Fig. 2). Evidently, this decrease is brought about by acoustic saturation of the (−) NMR mode of Mn^{55}.

DISCUSSION OF RESULTS

The character of the interaction of the acoustic oscillations with the nuclear spin system depends on the form of the nuclear spin-phonon coupling. For nuclei of magnetic atoms, the interaction of the nuclear spins with the vibrations of the lattice via electron spin waves is dominant (the magnetoelastic mechanism). The effect of this mechanism in cubic antiferromagnets was considered by Fedders[10] and by Merry and Bolef[9]. We write the effective Hamiltonian of the interaction of the nuclear spin with the sound in the form

$$\mathcal{H}_{\text{int}}=\hbar\gamma_N H_{1a} I=\tfrac{1}{4}\hbar\gamma_N H_{1a}(I_+e^{-i\omega t}+I_-e^{i\omega t}), \qquad (1)$$

where I is the nuclear spin, H_{1a} is the alternating magnetic field produced at the nucleus by the ultrasonic oscillations, γ_N is the nuclear gyromagnetic ratio, and ω is the sound frequency. The exprssion for this field is determined by the form of the spin-phonon coupling. In the case of the magnetoelastic mechanism, the presence of ultrasonic deformations leads to an effective magnetoelastic field H_{ME}:[9]

$$\mathbf{H}_{ME}=\nabla_M E_{ME}=\nabla_M\left(\frac{B_1}{M_0^2}\sum_{i=1}^{3}M_i^2 e_{ii}+\frac{B_2}{M_0^2}\sum_{i>j}M_i M_j e_{ij}\right), \qquad (2)$$

where M_i is the ith component of the magnetization of the sublattice, M_0 is the saturation magnetization of the sublattice, B_1 and B_2 are magnetoelastic constants, and $e_{\alpha\beta}$ is the $\alpha\beta$ component of the deformation tensor. However, the nuclei "feel" the amplified alternating field H_{1a} due to magnetoelastic modulation of the large hyperfine field

$$H_{1a}(\pm)=H_N \operatorname{tg}\beta=2AS\gamma_e\omega_E/\hbar\gamma_N\omega_{e\pm}^2, \qquad (3)$$

where β is the angle of rotation of the magnetization M_0, γ_e is the electron gyromagnetic ratio, ω_E is the exchange frequency, A the hyperfine constant, S the

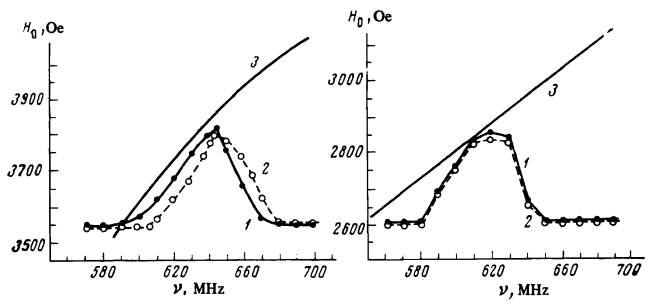

FIG. 3. Dependence of the field H_0 for AFMR in $RbMnF_3$ (a) and $KMnF_3$ (b) on the sound frequency ν. Curves 1, 2–experimental curves for the case of acoustic powers of 3×10^{-3} and 2×10^{-3} W/cm^2, respectively in the sample, 3–theoretical curve.

electron spin, $\omega_{e\pm}$ the frequencies (+) and (−) of the antiferromagnetic modes, and $B_\pm$ the magnetoelastic constant for these modes: the coefficient Γ determines the angular dependence of the magnetoelastic interaction,

$$(\mathbf{H}_{ME}\varepsilon_\pm)^2=B_\pm^2\Gamma_\pm e_{ij}^2/M_0^2,\ \varepsilon_+=\mathbf{H}_0/H_0,$$
$$\varepsilon_-=[\mathbf{M}_0\mathbf{H}_0]/M_0H_0. \tag{4}$$

Because of the large H_N and small H_A, the electronic and nuclear modes are strongly coupled, and one can speak only of the electron-like and nuclear-like frequencies. The equation for the resonance frequencies has the following form:[3]

$$(\omega^2-\omega_{e1,2}^2)(\omega^2-\omega_N^2)-\omega^2\omega_T^2=0, \tag{5}$$

where $\omega_{e1,2}$ are the frequencies of the AFMR unperturbed by interaction with the nuclear spins; $\omega_N = \gamma_N H_N$, $\omega_T = 2\gamma_e\sqrt{H_E H_{NE}}$, H_E is the exchange field; $H_{NE} = 9.43/T_N$ is the field of nuclear polarization on the electron spin; T_N is the effective nuclear spin temperature. For $\omega_N^2 \ll (\omega_e^2 + \omega_T^2)$, the approximate solutions of Eqs. (5) have the form

$$\omega_{e-}^2\approx\omega_{e1}^2+\omega_T^2, \tag{6a}$$
$$\omega_{e-}^2\approx\omega_{e2}^2+\omega_T^2, \tag{6b}$$
$$\omega_{n+}^2\approx\omega_N^2(1-\omega_T^2/\omega_{e+}^2), \tag{6c}$$
$$\omega_{n-}^2\approx\omega_N^2(1-\omega_T^2/\omega_{e-}^2), \tag{6d}$$

where $\omega_{e\pm}$ and $\omega_{n\pm}$ are the electron-like and nuclear-like frequencies.

When the field H_0 is parallel to the [001] axis, the frequencies $\omega_{e\pm}$ have the following form for configurations with spin flip:

$$\omega_{e+}^2=\gamma_e^2(H_0^2-{}^5/_2H_EH_A+\omega_T^2/\gamma_e^2), \tag{7a}$$
$$\omega_{e-}^2=\gamma_e^2(3H_EH_A+\omega_T^2/\gamma_e^2) \tag{7b}$$

for $RbMnF_3$[8] and

$$\omega_{e+}^2=\gamma_e^2\left[\left(H_0+\frac{K_2}{M_0}\right)\left(H_0+4\frac{K_2}{M_0}\right)+2\lambda(K_1-K_4)+\frac{\omega_T^2}{\gamma_e^2}\right], \tag{8a}$$

$$\omega_{e-}^2=\gamma_e^2\left[\left(H_0+\frac{K_2}{M_0}\right)\frac{K_2}{M_0}-4\lambda K_4+\frac{\omega_T^2}{\gamma_e^2}\right] \tag{8b}$$

for $KMnF_3$,[7] where K_1, K_2 and K_4 are the constants of the axial, orthorhombic, and cubic anisotropies,[11] and λ is the exchange constant.

The term ω_T, which governs the coupling of the unperturbed electron and nuclear modes, is inversely proportional to the temperature of the nuclear spin system. While this term is small at high temperatures ω_T is of the order of ω_e at low temperatures and the electron and nuclear modes become strongly coupled. As a result, significant low-temperature shifts appear in the electron and nuclear frequencies. In our experiments, the temperature of the nuclear spin system changes because of the applied acoustic field and in the final analysis, it leads to a shift in the resonance field H_0 for AFMR.

The shift in the field for AFMR is observed when the sound frequency falls in the range of frequencies which begins with the nuclear-like resonance frequency ω_{n+} (at a temperature $T = 4.2°K$) and extends to the hyperfine frequency ω_N. The change in the shift as a function of the frequency of nuclear pumping is well described by Eqs. (6c) and (7a) for $RbMnF_3$ and (6c), (8a) for $KMnF_3$. As nucleation centers of the saturation, one has the inhomogeneities in the crystal, near which the nuclei resonate at a frequency close to ω_N.[7] When the acoustic power reaches some threshold value (of the order of 0.4×10^{-3} W/cm^2), saturation of the entire

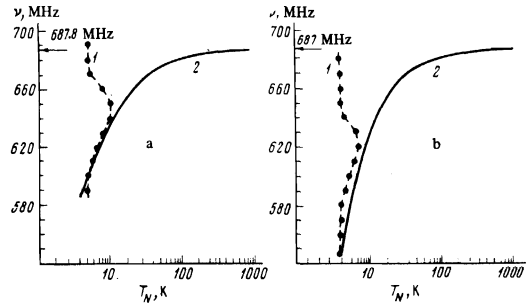

FIG. 4. Frequency of the acoustic pump ν as a function of the effective nuclear temperature T_N for $RbMnF_3$ (a) and $KMnF_3$ (b). Curve 1 was calculated from the experimental data obtained at constant acoustic power; 2–theoretical curve.

sample takes place at the pumping frequency. We calculated the effective temperature T_N of the nuclear spin system as a function of the pump frequency (Fig. 4). It was established that T_N increases to very large values on a significant increase in the pump power, when the sound frequency approaches ω_N. It is possible that the saturation of the nuclear spin system is due to the excitation of nuclear spin waves, which have a frequency spectrum which includes the range of frequencies indicated.[6].

To determine the magnetoelastic coupling constants from (4), it is necessary to find the values of H_{1a} and e_{ij}. The field H_{1a} can be estimated from the identical shifts of the AFMR by electromagnetic (em) and acoustic (a) fields ($H_{1a} = H_{1em}$):

$$H_{1em}=H_1^0\eta,\ \eta=2^{-1/2}H_N(H_0+H_A)(\gamma_e/\omega_{e+})^2, \tag{9}$$

where H_1^0 is the radiofrequency field in the circuit and η the gain of the field in the antiferromagnet.[7]

At $H_1^0 \sim 0.1$ Oe, $H_{1em} \sim 15.5$ Oe for $RbMnF_3$ and 11.4 Oe for $KMnF_3$. It must be noted that the values of H_{1a} that we have found satisfy the saturation condition of the nuclear spin system:

$$\gamma_N H_{1a} T_1 T_2 \geqslant 1, \tag{10}$$

where T_1 and T_2 are respectively the longitudinal and transverse relaxation times of the nucleus.

The amplitude of the deformation was measured by a capacitative pickup[12] and was also calculated from the voltage U on the piezoelectric transducer and the value of the electromechanical coupling coefficient ρ, $e_{ij} = 2\sqrt{2}\rho U/vZ$, where Z is the acoustic impedance of the sample and v the sound velocity. Both methods give the same values accurate to within 20%.

The magnetoelastic constant B is determined from relations (3) and (4):

$$B=\left(\frac{\hbar\gamma_N\omega_{e+}^2M_0}{2AS\gamma_e\omega_E}\right)\frac{H_1}{e\sqrt{\Gamma}}=\frac{\gamma_N\omega_{e+}^2M_0H_1}{2\gamma_e\omega_N\omega_Ee\sqrt{\Gamma}}. \tag{11}$$

The angular coefficient in (10) is

$$\Gamma=\frac{(1-3\cos^2\psi)\sin^2 2\psi}{3\cos^4\psi-10\cos^2\psi+3}$$

$\psi \approx 90°$ (ψ is the angle between H_0 and k).[9] Let us evaluate expression (11), assuming that for identical shifts of AFMR, $H_{1em} = H_{1a}$, and taking the following values of the parameters: $\Gamma \approx 5\times10^{-1}$; $e \approx 10^{-7}$, $M_0 \approx 300$ Oe, $\omega_{e+} = 9.14$ GHz, $\omega_E(RbMnF_3) = 1.57\times10^{12}$ Hz, $\omega_N(KMnF_3) = 687.8\times2\pi$ MHz, $\omega_E(KMnF_3) = 1.58\times10^{12}$ Hz, and $\omega_N(KMnF_3) = 687\times2\pi$ MHz. Then the respective magnetoelastic constants are $B(RbMnF_3)$

$= 1.3 \times 10^6$ erg/cm^3, $B(KMnF_3) = 1.2 \times 10^6$ erg/cm^3. Comparison of the values given above for the magnetoelastic constants with the values ($B(RbMnF_3) = 2.1 \times 10^6$ erg/cm^3 [9] and $B(KMnF_3) = 10^6$ erg/cm^3 [13]) obtained by the method of direct acoustic resonance indicates satisfactory agreement of the results. Evidently some divergence results from the inaccuracies of measurement of the magnetic damping of the sound by the method of direct ANMR.

In the case of the method of calculation of H_{1a} given above, the equivalence of the threshold values of the fields H_{1a} and H_{1em} for the appearance of the AFMR shift has been established. For example, the values that we found for $KMnF_3$ were $H_{1a} = 2.5$ Oe and $H_{1em} = 2.23$ Oe.[7]

The experiments showed that our method is superior in its sensitivity and the possibilities that it offers for study of the dynamics of internal interactions in magnetic crystals to the method of direct ANMR. Evidently, the greatest advantages of the method of double resonance will appear in study of magnetically dilute materials and nuclei with relatively weak magnetoelastic coupling.

As a result of our studies by the method of double nuclear antiferromagnetic resonance, it has been established that the acoustic energy is transferred by magnetostriction to the electron spin system, and from it via hyperfine interactions to the spin system of the Mn^{55} nuclei. The acoustic saturation has been studied through the shift in the field for AFMR and the magnetoelastic constants have been determined for nuclei of Mn^{55} in $RbMnF_3$ and $KMnF_3$. Additional confirmation of the existence of dynamic coupling of the nuclear and electron spin systems has been obtained from the field shift of the AFMR at acoustic saturation.

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