NMR and the domain structure in $FeBO_3$ single crystals

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Experimental results are presented of an investigation of the field dependence of nuclear magnetic resonance in $FeBO_3$ at 77°K. The nature of the signal is discussed and a qualitative explanation of the hysteresis of the spin-echo amplitude is proposed. The possibility of measuring K/μ_s by the NMR technique is demonstrated.

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INTRODUCTION

Nuclear magnetic resonance (NMR) in the rhombohedral antiferromagnet $FeBO_3$ was investigated by a number of authors [1-3]. The results obtained on the temperature dependence of the NMR frequency were used for quantitative comparison with spin-wave theory for crystals of the easy-plane type. The hitherto investigated [1-3] $FeBO_3$ samples constituted a set of randomly oriented plates, and the authors did not succeed in identifying the observed resonance with excitation of nuclei in the domains or in the walls. Information obtained by measuring the NMR gain was insufficient to identify the signal.

We investigate in this paper the field dependences of NMR in $FeBO_3$ for the purpose of obtaining new experimental data aimed at clarifying the origin of the signal, and use the results to discuss the magnetization-reversal processes in such crystals.

PROCEDURE AND SAMPLES

The NMR was investigated with a pulsed spectrometer at a frequency 75.325 MHz, corresponding to the resonance of the $^{57}Fe^{3+}$ nuclei [3] at liquid-nitrogen temperature. The sensitivity of the receiving system ensured observation of spin echo from one $FeBO_3$ single crystal with the natural content of the ^{57}Fe isotope. The investigated samples were obtained by the method described in [4] and constituted six-faced plates, green in color, of thickness ~0.1 mm and area ~12 mm^2. Owing to the weak anisotropy of sixth order in the basal plane, the magnetizations in the sublattices were oriented along three directions parallel to twofold symmetry axes.

In the determination of field dependence, the earth's magnetic field was taken into account.

GENERAL RELATIONS FOR THE NMR GAIN IN $FeBO_3$

The influence of an external magnetic field on the NMR in the region of weak fields is determined essentially by the character of the change of the domain structure of the sample in the course of the magnetization. Since iron borate is transparent in the visible region of the spectrum, magnetooptical methods are used to study the domain structure. Investigations of this type were performed by a number of workers [5,6] who proposed a model according to which the $FeBO_3$ crystal is made up of domain layers with different directions of magnetization in the basal plane, and in each layer there are in turn 180° domains separated by Neel walls. When an external field is applied, mobility of the Neel walls is observed in fields smaller than 1 Oe. The boundaries between the layers are 90° [5] or 120° [6] Bloch walls.

Thus, one can expect NMR in $FeBO_3$ to be due either to nuclei contained in the domains or in the walls of both types. It is possible that several mechanisms contribute simultaneously to the enhancement of the NMR, as is the case, for example, in hematite [7]. Let us estimate the gain η_d due to the rotation of the magnetization in the domain.

The corresponding expressions for η_d were obtained by Anderson [8]

$$\eta_d \approx \left| \frac{H_{hff}\mu_s}{18K}\cos\varphi \right|, \quad H_0 = 0, \tag{1}$$

$$\eta_{d\parallel} \approx \left| \frac{H_{hff}\mu_s \cos\varphi}{\mu_s H_0 \sin\varphi + 18K\cos 6\theta} \right|, \quad H_0 \| H_1, \tag{2}$$

$$\eta_{d\perp} \approx \left| \frac{H_{hff}\mu_s \sin\varphi}{\mu_s H_0 \sin\varphi + 18K\cos 6\theta} \right|, \quad H_0 \perp H_1, \tag{3}$$

where H_0 is the external magnetic field, H_1 is the radio-frequency field, H_{hff} is the local field at the nucleus, θ is the angle between the easy axis of the crystal and the magnetization direction in the sublattice, φ is the angle between the applied field and the magnetization direction in the sublattice, μ_s is the electronic magnetization in the domain, and K is the magnetic crystallographic anisotropy constant.

If only an alternating field is applied to the sample, then according to (1) we have at $\varphi = 0$

$$\eta_d \approx |H_{hff}\mu_s/18K| \approx 7 \cdot 10^3 \tag{4}$$

for H_{hff} = 546 kOe and 18 K/μ_s = 8 Oe. [6]

In the case of excitation of nuclei in a 180° wall we have in accordance with [9]

$$\eta_w \approx \frac{2CH_{hff}\mu_s \sin\psi}{M\delta[(\omega_0^2 - \omega_{NMR}^2) + (\beta/M)^2\omega_{NMR}^2]}. \tag{5}$$

where C is a constant that depends on the type of wall, ψ is the angle between the electron spin and the easy axis, M is the mass of the wall, δ is the width of the wall, β is the damping parameter in the Landau-Lifschitz equation, and ω_0 is the resonant frequency of the wall oscillations. The maximum gain $(\eta_w)_{max}$, following (5), is observed in the case when $\omega_0 = \omega_{NMR}$. At the center of the wall we then have at C = 1

$$(\eta_w)_{max} \approx H_{hff}\gamma_0 H_{res}/\omega_{NMR}\Delta H, \tag{6}$$

where the damping parameter β is expressed in analogy with [9] in terms of the width ΔH of the ferromagnetic resonance line. Using the value $\gamma_0 H_{res}$ = 34.5 GHz and ΔH = 20—100 Oe at T = 77° K [10], we obtain $(\eta_w)_{max}$ ≈ 2.5×10^6—12.5×10^6.

In spin-echo experiments, the given value of η_w decreases by a factor [9] $s\delta/v$, where s is the area of the wall and v is the volume of the sample. In view of the

lack of experimental data, it is impossible to estimate the average value of η_w. Thus, the estimates of the gains show that there are several mechanisms that lead to η in the interval 10^4—10^6. An external magnetic field changes the state of the domain structure of the sample and accordingly can greatly influence the NMR signal.

EXPERIMENTAL RESULTS AND DISCUSSION

We present below the experimental data for two cases of a field dependence of the NMR, when the constant field is parallel to the basal plane of the crystal and in one case is parallel to H_1 and in the other case perpendicular to H_1. Figure 1 shows the corresponding plots of $A/A_m(H_0)$, and also the hysteresis loop of the sample.

In fields stronger than $H_{sat} \approx 16$ Oe, the sample is in a one-domain state and the observed signal (Fig. 1c) is connected with excitation of nuclei inside the domain. For $H_1 \parallel H_0$ at $H_0 > H_{sat}$ we have in accordance with (2) $\eta_{d\parallel} = 0$, since $\varphi = 90°$ and spin echo should not be observed (Fig. 1b). Proof of excitation of nuclei in the domain can be obtained by comparing the NMR gain calculated in the presence of an external field in accordance with (3) and the measured gain.

According to (3) we have

$$\eta_{d\perp} \approx \left| \frac{H_{hff}}{H_0 - 18K/\mu_s} \right| \approx 2.3 \cdot 10^4 \qquad (7)$$

for $H_0 = 2H_{sat} \approx 32$ Oe and $\theta = 30°$. To measure $\eta_{d\perp}$, we plotted the function $A(H_1)$ under the assumption that the maximum spin-echo amplitude corresponds to the condition

$$\gamma \eta_{d\perp} H_1 \tau = 2\pi/3, \qquad (8)$$

where γ is the gyromagnetic ratio and τ is the duration of the RF pulses. The value obtained in this manner is of the order of 1.5×10^4 and is close to that calculated by formula (7).

In the absence of an external field it is difficult to point unambiguously to the enhancement mechanism. In any case, there are no grounds whatever for neglecting the influence of the walls, and it is possible that those

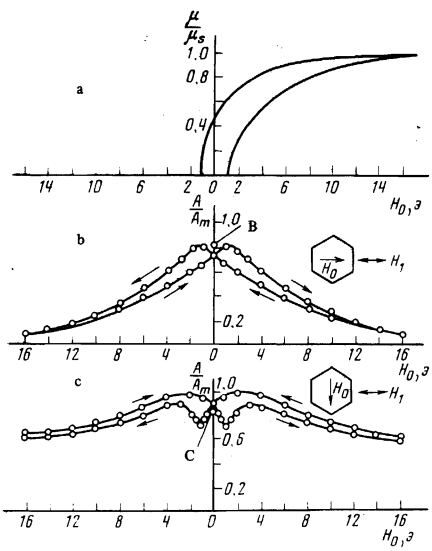

FIG. 1. Hysteresis loop (a) and dependence of the relative amplitude of the spin echo on the field for $H_0 \parallel H_1$ (Fig. b) and for $H_0 \perp H_1$ (c) for the reversal of the magnetization of single-crystal $FeBO_3$. A_m is the maximum echo amplitude.

nonlinear effects which are mentioned in [2] (the dependence of the relaxation times and the waveform of the echo on H_1) and which are observed in our experiments are determined by this singularity of the resonance in $FeBO_3$. Taking the foregoing into account, we chose the largest value of H_1 when plotting the field dependences in Figs. 1b and 1c, and since the average $\eta_d < \eta_w$, the main contribution to the NMR intensity was made in this case by nuclei located inside the domains.

We proceed to consider the singularities of NMR in the case of reversal of the sample magnetization. A decrease of the magnetizing field $H_0 < H_{sat}$ leads to the appearance of an NMR signal in the case $H_0 \parallel H_1$ (Fig. 1b). The latter may be due to the appearance of the projection $\mu_s \perp H_1$ responsible for the excitation of the nuclei inside the domains, or to the appearance of walls parallel to H_1. Taking into consideration data obtained by others [5,6] and the NMR results in Figs. 1b and 1c, we present a model describing the reversal of magnetization in $FeBO_3$. The formation of the domain structure in fields $H_0 < H_{sat}$ starts with the formation of domain layers separated by Bloch walls in the thickness of the crystal (Fig. 2a). The orientation of the magnetization vectors in these layers changes relative to the field direction towards the closest easy-magnetization directions, and the spin rotation in the walls takes place around an axis perpendicular to the basal plane of the sample. With further decrease of the field, 180° domains separated by Neel-type walls start to appear inside each layer (Fig. 2b). In these fields, the magnetization reversal is effected simultaneously by rotation of the magnetization in the domain layers and by displacement of the Neel walls; the subsequent change in the domain structure is shown in Fig. 2c. The displacement of the walls is accompanied by easily observed Barkhausen jumps, and the rotation of the magnetization in the domain layers is confirmed by data obtained by the microwave procedure [11]. At $H_0 = 0$, the described state of the domain structure is preserved, and the larger value of the spin echo at $H_0 \perp H_1$ is explained by the fact that the projection of the remanent magnetization, which is perpendicular to H_1, is in this case larger than at $H_0 \parallel H_1$.

When the direction of the magnetizing field is reversed, a maximum of the NMR signal amplitude is observed for $H_0 \parallel H_1$, and a minimum for $H_0 \perp H_1$ (Figs. 1b and 1c) in fields on the order of the coercive force. This is due to the change of the contribution of the nuclei inside the domains to the NMR intensity, a change due to rotation processes that lead in one case to an increase

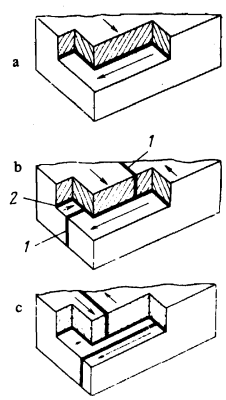

FIG. 2. Schematic model of reversal of $FeBO_3$ magnetization. The arrows show the direction of the magnetization in the domain. In Fig. b, 1 and 2 are the Neel and Bloch walls, respetively.

of $\mu_S \perp H_1$ and in the other case to a decrease. For the same reason, the spin-echo amplitude in the demagnetized state (points B and C in Figs. 1b and 1c) differ from the corresponding values in the remanent-magnetization state.

In conclusion, we call attention to the fact that the signal from the nuclei in the domains that is observed in a field $H_0 > H_{sat}$ can be used to measure the value of K/μ_S in $FeBO_3$. Thus, it follows from (3) that at $\varphi = 90°$ we have

$$\frac{\eta_{d\perp}(\theta=30°)}{\eta_{d\perp}(\theta=0°)} = \frac{H_0+18K/\mu_s}{H_0-18K/\mu_s}.$$

Since $A \sim \mu_d$, it suffices to measure the ratio of the spin-echo amplitudes at two orientations of the crystal ($\theta = 0°$ and $\theta = 30°$) in a one-domain state in order to calculate K/μ_S. At $H_0 = 32$ Oe, the indicated ratio is $A(30°)/A(0°) = 1.5$, corresponding to $18\,K/\mu_S \approx 6.4$ Oe; the latter agrees with [6].

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