Magnon relaxation in antiferromagnetic $FeBO_3$ at low temperatures

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Low-temperature relaxation of magnons in the antiferromagnet $FeBO_3$ is investigated at pumping frequencies $\omega_p/2\pi = 26.2$ and 35.4 GHz and temperatures between 1.2 and 20 K. The temperature and field dependences of the magnon relaxation parameter $\Delta\omega_{\mathbf{k}}$ indicate that the main contribution to $\Delta\omega_{\mathbf{k}}$ for $1.2 \leqslant T \leqslant 4.2$ K is from three-particle magnon-phonon interactions. The characteristic magnon-phonon interaction energy is found to be $\Theta \approx 10^{-14}$ erg. It is suggested on the basis of the data obtained that magnon-phonon scattering should be enhanced at low temperatures by magnetic impurity ions present in the crystal.

Much theoretical and experimental work has been done recently on parametric excitation of magnons in antiferromagnets ($CsMnF_3$, $MnCO_3$, $FeBO_3$, $CuCl_2 \cdot 2H_2O$, etc.) containing various magnetic ions.[1–5] Most of this work has been done on antiferromagnets with an "easy-plane" anisotropy, because their magnon spectrum has a quasi-acoustic branch which lies in the experimentally accessible microwave region. The parametric decay of a microwave phonon of frequency ω_p into two magnons with frequency $\omega_p/2$ and wave vectors **k** and $-$**k** has a threshold, and the threshold field h_c at the specimen is proportional to the relaxation time of the excited magnons. Much of the interest in such studies derives from the fact that they enable one to directly analyze how the magnon relaxation time depends on $\omega_{\mathbf{k}}$, **k**, and on the external conditions (temperature T, magnetic field, etc.). The results can then be compared with theory to identify the principal interaction processes both within the magnon system and between the magnons and other types of elementary excitations in the crystal (phonons, nuclear magnons, fluctuations in the nuclear magnetization) and with crystal defects (impurities, boundaries). As usual, interactions that occur in perfect crystals and the associated relaxation processes will be said to be intrinsic, while interactions involving defects will be called nonintrinsic. We note that these interactions also determine the stationary threshold state for the parametrically excited spin system, and in particular the magnon spectrum and the mechanism limiting the number of magnons that are generated.

Our experimental results show that as the temperature decreases, there is an abrupt falloff in the contribution to the magnon relaxation from the magnon-magnon interactions. In most cases, the dominant interaction here is a three-magnon process in which a parametric magnon coalesces with a thermal magnon in the quasi-acoustic branch to generate a magnon in the quasi-optical branch. The relaxation time for this three-magnon interaction thus depends exponentially on T down to temperatures corresponding to the energy gap in the spectrum of the quasi-optical branch of the magnon spectrum. For four-magnon processes, the relaxation time is proportional to T^n with a large exponent $n \geqslant 2$. At low temperatures, the magnon relaxation should thus be governed by the interaction either with other quasiparticles or with crystal defects.

We note that since weak interactions in antiferromagnets are generally enhanced by exchange effects, the temperature at which the the magnon-magnon relaxation ceases to be a dominant factor may be relatively high (of the order of a few degrees Kelvin).

The theory predicts that among the intrinsic channels for low-temperature magnon relaxation, scattering by thermal phonons may play a leading role. We will show below that the decrease in the magnon-phonon scattering probability as T decreases is considerably slower than for magnon-magnon scattering.

The magnon-phonon interaction alters the static and high-frequency magnetic properties of antiferromagnets, and many of these effects have been studied previously (in particular, the magnetostriction, the magnetoelastic energy gap in the magnon spectrum, and the dependence of the speed of sound on the magnetic field). The experimental and theoretical work on the effects of magnon-phonon interaction on the static and linear dynamic properties of antiferromagnets is reviewed in detail in Ref. 6.

Experiments on parametric excitation of magnons have revealed that the magnon relaxation is anomalous in the region where the magnon and phonon spectra intersect. Stimulated decay of a parametrically excited magnon into a magnon and low-energy phonon has been observed, as well as coalescence of two parametric magnons to generate a phonon. In addition, the magnon-phonon interaction has been found to alter the state of the parametrically excited spin system for fields above the threshold value. The influence of the magnon-phonon interaction on the nonlinear dynamics of antiferromagnets was reviewed in Ref. 7.

In our present work we attempted to detect a magnon-phonon contribution to the magnon relaxation in an antiferromagnet with an easy-plane magnetic anisotropy by analyzing the magnon relaxation in detail at low temperatures. We chose $FeBO_3$ as the easy-plane antiferromagnet (Neel temperature $T_N = 348$ K) because its large magnetoelastic and exchange relative to the magnon-magnon relaxation (see below). In addition, relaxation involving the interaction of electron magnons with the nuclear magnetic moments should be negligible for $FeBO_3$, because the principal isotope Fe^{56} has zero nuclear spin, while the concentration of Fe^{57} (with $S = 5/2$) is just $\sim 2\%$ in natural crystals. Furthermore, the magnetic and elastic properties of $FeBO_3$ have been investigated in detail. We refer to Ref. 8 for a summary of the main results and review of the literature.

The $FeBO_3$ crystal belongs to the D_{3d}^6 symmetry class.

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The spectra for magnons and phonons interacting in easy-plane antiferromagnets depend essentially on the ratio s/c of the strong-field limit of the magnon and phonon propagation velocities. For $FeBO_3$, $s > c$ and the magnon spectrum for a two-sublattice antiferromagnet has two branches which are given by[9,10]

$$(\omega_{1km}/\gamma)^2 = H(H+H_O) + H_\Delta^2 + \alpha_\parallel^2 k_\parallel^2 + \alpha_\perp^2 k_\perp^2, \quad (1)$$

$$(\omega_{2km}/\gamma)^2 = 2H_A H_E + H_{\bar{H}}(H+H_O) + \alpha_\parallel^2 k_\parallel^2 + \alpha_\perp^2 k_\perp^2 \quad (2)$$

for $d^{-1} \ll |\mathbf{k}| \ll \pi/a_0$, where d is the diameter of the crystal and a_0 is the lattice constant; ω_{ikm} and $\mathbf{k}$ are the circular frequency and the wave vector of the magnon; γ is the magnetomechanical ratio ($\gamma = 2\pi \cdot 2.8$ GHz/kOe for $FeBO_3$); H_E, H_D, and H_A are the exchange, Dzyaloshinskiĭ, and uniaxial anisotropy fields; the spectral parameter H_Δ^2 allows for the magnetoelastic interaction (among others); $\alpha_\parallel$ and $\alpha_\perp$ are the exchange constants, where $\parallel$ and $\perp$ indicate the direction relative to the principal axis of the crystal. For simplicity, we have omitted from (1) the terms corresponding to the dipole-dipole interaction and the anisotropy in the base plane.

The distortion of the phonon spectrum by the magnetoelastic interaction depends on the polarization and direction of propagation of the phonons. When the magnetic field $\mathbf{H}$ lies in the base plane of the crystal ($z \parallel C_3$, $x \parallel \mathbf{H}$), the distortion is greatest for phonons that move along the C_3 axis and are polarized along $\mathbf{H}$; in this case we have[11,12]

$$\omega_{ph}(k) = c_{tz}(1 - \gamma^2 \lambda_4^2 l_0^3 H_E/\mu_5 \omega_{1km}^2)^{1/2} k, \quad (3)$$

where $c_{tz} = (c_{44}/\rho)^{1/2}$ is the corresponding speed of sound in the limit $H \to \infty$ (for acoustic waves with the same polarization and direction); $\mu_5 = c_{44}/2$ is the elastic modulus; $\lambda_4 l_0^2 = 2B$ [14] is a component of the magnetostriction tensor; $M_0 = l_0/2$ is the magnetization of the sublattice.

Since the interpretation of the experimental data depends on the formulas used to describe the various contributions to the magnon relaxation parameter, we will derive our own expressions for the principal magnon relaxation channels in $FeBO_3$ (this is necessary because the published theoretical results differ greatly for certain processes, including magnon-phonon scattering). We consider only low temperatures $\varepsilon_{1k} \lesssim k_B T \ll \varepsilon_{20}$.

Sobolev[13] has calculated the relaxation parameter associated with three- and four-magnon interactions; the three-magnon interactions are limited to a single process in which two low-energy (quasi-acoustic) magnons merge to form a magnon in the upper (quasi-optical) branch (other three-magnon interactions are forbidden because they would violate conservation of energy and quasimomentum). Specializing to our situation, we can rewrite the expression for $\Delta\omega_{3m}$ in Ref. 13 as

$$\Delta\omega_{3M} = \frac{k_B T}{16\pi\hbar a}\left(\frac{\mu\tilde{H}}{\Theta_N}\right)^2\left(\frac{J_0}{\Theta_N}\right)\left(\frac{\varepsilon_{20}}{\varepsilon_{10}}\right)^4 [(1+a^2)\,\mathrm{sh}\,ab$$
$$-2a\,\mathrm{ch}\,ab]\exp(-b)\left[1-\exp\left(-\frac{\varepsilon_{1k}}{k_B T}\right)\right], \quad (4)$$

where

$$a = \left[1-\left(\frac{\varepsilon_{10}}{\varepsilon_{1k}}\right)^2\right]^{1/2}, \quad b = \frac{\varepsilon_{20}^2 \varepsilon_{1k}}{2\varepsilon_{10}^2 k_B T},$$

$$\tilde{H} = H + 2H_D\left(\frac{H+H_D}{H_E}\right)^2,$$

$$\Theta_N = \frac{\hbar s}{v_0^{1/3}}, \quad J_0 = \frac{\mu H_E}{S},$$

$s = 2\pi a\gamma$ is the strong-field limit of the magnon propagation velocity, v_0 is the unit cell volume, and S is the spin ($S = 5/2$ for $FeBO_3$). Formula (4) shows that $\Delta\omega_{3m}$ decreases exponentially with T at low temperatures.

Many four-magnon processes can occur in fields that are weak compared with the Dzyaloshinskiĭ field (since $H_D = 108$ kOe for $FeBO_3$, this condition always holds in our case); according to Ref. 13, for $k_B T \ll \varepsilon_{1k}$ the most important of these are the pure four-body processes in which a pair of magnons in the quasi-acoustic branch coalesce to give two magnons in the same branch. The expression for $\Delta\omega_{4m}$ in this case is[13]

$$\Delta\omega_{4M} = \frac{J_0}{16(2\pi)^3\hbar}\left(\frac{J_0}{\Theta_N}\right)\left(\frac{\varepsilon_{10}}{\Theta_N}\right)^3\left(\frac{k_B T}{\Theta_N}\right)^2 f\left(\frac{\varepsilon_{1k}}{k_B T}, \frac{\varepsilon_{10}}{k_B T}\right), \quad (5)$$

where f is a function with values ~ 1.

Other processes involving the interaction of the electron spins with phonons or with nuclear spins may also contribute to magnon relaxation in a perfect crystal. We will not consider magnon relaxation involving the nuclear spins, since we have already noted that it is negligible in $FeBO_3$ crystals that have not been enriched with Fe^{57} isotope.

Relaxation involving magnon-phonon interactions is dominated by two processes: coalescence of a parametric magnon with a thermal phonon to form another magnon, and decay of a parametric magnon into a magnon and a phonon (or into two phonons). It follows from general considerations that the decay of a magnon into two phonons contributes little to the magnon relaxation, because the contribution is proportional to the square of the small magnetoelastic coupling parameter. The total relaxation parameter for the first two processes was calculated in Refs. 14–17 for the case $\delta > 1$, $\varepsilon_{1k} \ll \delta k_B T$ of interest to us ($\delta = s/c$); the result is[17]

$$\Delta\omega_{mp} = \frac{2\delta^2 k_B}{3\pi\hbar}\left(\frac{\Theta^2}{Mc^2}\right)\frac{J_0^2 T}{\Theta_N^3} f(\hbar s k/\varepsilon_{1k}), \quad (6)$$

where

$$f(x) = \frac{4}{\delta^2-1} - \ln\left[1 + \frac{4\delta^2}{(\delta^2-1)^2}(1-x^2)\right], \quad \delta^{-1} < x \leqslant 1, \quad (7)$$

$$f(x) = \frac{4\delta x}{\delta^2-1} - \ln\left[\frac{1+2\delta x/(\delta^2-1)}{1-2\delta x/(\delta^2-1)}\right], \quad 0 \leqslant x < \delta^{-1}, \quad (8)$$

$M = \rho v_0$ is the mass of the unit cell, $\Theta = v_0 B_i$ is the characteristic magnetoelastic interaction energy, and B_i is expressible in terms of the components of the magnetostriction tensor. The result for $\Delta\omega_{mp}$ in Ref. 13 differs from (6) by a numerical factor (it is 50% larger), evidently because the parameter B_i (and hence Θ) is not uniquely determined, The expression $B_i = |B_{11} - B_{12}|/2$ was used in Ref. 13.

Comparison of expression (4)–(6) indicates that the magnon-phonon interaction dominates the intrinsic magnon relaxation processes at low temperatures, as stated in the Introduction. This is because its low-temperature contribution to the relaxation is proportional to T as T decreases and thus falls off more slowly than the relaxation parameters for the other processes.

Expressions (7) and (8) can be simplified if δ is sufficiently large ($\delta \approx 3$ for $FeBO_3$). We than find from (7) that there is a wide range of fields $0 < H < (1-\delta^{-2})H_0$ for which $f(x) \approx 4\delta^2 x/(\delta^2-1)^2$ and

$$\Delta\omega_{mp} \infty skT/\varepsilon_{1k} \tag{9}$$

(here H_0 is the field at which magnons with $k=0$ are excited).

EXPERIMENTAL METHOD AND SPECIMENS

We used a direct amplification spectrometer (described in detail in Ref. 18) to investigate parametric magnon excitation. The TE_{011} and TE_{012} modes were excited in a high-Q cylindrical resonator ($Q \sim 10^4$). A roll of cigarette paper secured the crystal to the bottom of the resonator at an antinode of the pumping microwave field **h**, which was parallel to **H** and perpendicular to the C_3 axis of the crystal. This arrangement avoided the generation of elastic strain in the crystal during cooling (elastic strain can appreciably alter the magnon spectrum in $FeBO_3$).

Continuous microwave oscillators were used to pump the system at the two frequencies $\omega_p/2\pi = 35.4$ and 26.2 GHz. Long pumping pulses were used in some of the measurements; the pulse length ranged from 0.1 to 10 ms and the repetition rate was 50 Hz.

The microwave pulse transmitted through the resonator containing the crystal was detected and displayed on an oscilloscope. The amplitude of the pulse as a function of the static magnetic field was recorded by an X, Y plotter. The relative field strengths h at the crystal were measured in a series of experiments by a square-law detector whose output voltage was proportional to h^2 when the detector was weakly coupled to the output of the resonator. The known resonator parameters and microwave input power were then used to calculate the absolute field strength h from the standard formulas for the field distribution in the resonator (the microwave power was measured by a thermistor). The absolute error in measuring h at the crystal was $\sim 20\%$ (the relative error $\sim 3\%$ was considerably smaller).

It was shown in Ref. 8 that parametric magnon excitation in $FeBO_3$ has a sharply defined threshold at temperatures $\lesssim 7$ K, and there are two critical fields h_{c1} and h_{c2} ($h_{c1} > h_{c2}$) at which parametric excitation begins and terminates, respectively. The microwave power absorbed by the crystal is discontinuous at $h = h_{c1}$ or h_{c2}. Since h_{c1}/h_{c2} may be as large as 10 in $FeBO_3$ at low temperatures, two

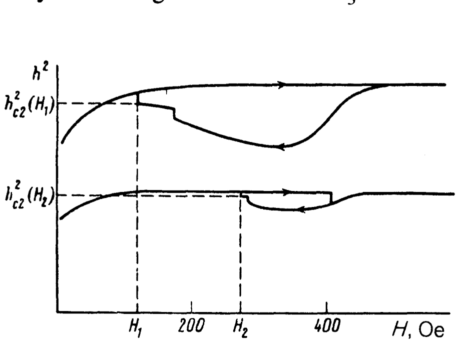

FIG. 1. Trace of the microwave power $P_{trans} \propto h^2$ transmitted by the resonator as a function of the magnetic field H.

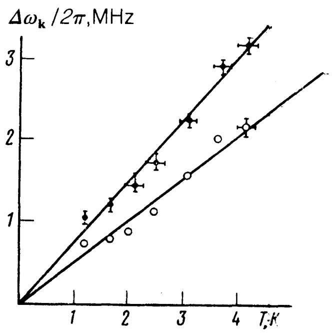

FIG. 2. Relaxation parameter $\Delta\omega_k$ versus temperature for $\omega_p/2\pi = 35.4$ GHz; ●, $H = 200$ Oe; ○, $H = 300$ Oe.

different techniques were needed to measure the critical fields. The field h_{c1} was found by noting the field strength at which the amplitude of the microwave pulse from the resonator suddenly dropped; the errors in h_{c1} and h were essentially the same. We found h_{c2} by recording $P_{trans}(H)$, the microwave power transmitted through the resonator, on a plotter while the resonator was pumped continuously at a constant incident power P_{inc} (see Fig. 1).[19] Some of the $\sim 10\%$ error in measuring h_{c2} (the field at which P_{trans} suddenly increased) was due to power fluctuations in the microwave oscillator while the field H was swept.

In the measurements for $T = 1.2$–4.2 K, the resonator was filled with liquid helium in order to cool the crystal more effectively. The temperatures in this range were measured to better than ± 0.05 K by measuring the saturated helium vapor pressure. A semiconductor thermometer was used at higher temperatures, and in this case the measurement error was considerably larger (± 0.5 K) because the heat transfer occured through helium vapor.

The specimens were naturally faceted single-crystal wafers with cross section $\sim 2\times 2$ mm^2 and thickness 0.5–2 mm. Depending on the thickness, their color ranged from green to nearly black. The plane of the wafers coincided with the base plane of the crystal.

EXPERIMENTAL RESULTS

Parametric excitation of magnons in antiferromagnets with an easy-plane anisotropy and weak ferromagnetic properties was analyzed theoretically in Ref. 20. If $\omega_p \ll \omega_{20}$ holds and **h**, **H**, and C_3 are mutually orthogonal, the threshold field h_c is related to the relaxation parameter $\Delta\omega_k$ of the excited magnons by

$$h_c = \omega_p \Delta\omega_k/\gamma^2(2H + H_D). \tag{10}$$

Existing theory suggests that the abrupt threshold for parametric excitation results from turning off a component of the relaxation. Although this component has yet to be identified conclusively, experimental data (see, e.g., Ref. 3) suggest that the nonintrinsic magnon relaxation mechanism involving paramagnetic impurity ions becomes saturated (we reached this conclusion previously in Ref. 18). Since we were interested in the intrinsic relaxation mechanisms we therefore studied the time-independent component of the relaxation $\Delta\omega_k$, which is determined by the threshold field h_{c2}.

The temperature dependence of the relaxation parameter $\Delta\omega_k$ calculated by (10) is shown in Fig. 2; we see that $\Delta\omega_k \propto T$ to within the experimental error.

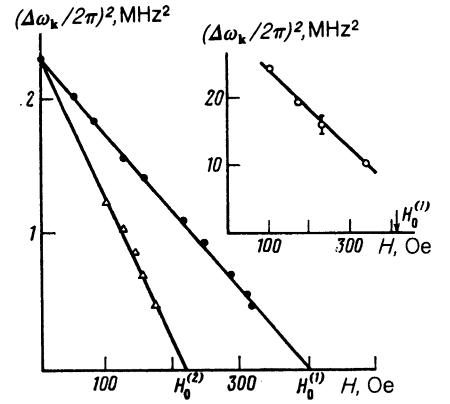

FIG. 3. Field dependence of the relaxation parameter $\Delta\omega_k(H)$: ●, $\omega_p/2\pi = 35.4$ GHz, $T = 1.2$ K; △, 26.2 GHz, 1.2 K; O, 35.4 GHz, 18 K.

The dependence of $\Delta\omega_k(H)$ for the two pumping frequencies is shown in Fig. 3. The fields $H_0^{(1)}$ and $H_0^{(2)}$ found by extrapolating the curves $\Delta\omega_k(H)$ to $\Delta\omega_k = 0$ agree with the values H_0 found by Eq. (1) if one takes $H_\Delta^2 = 0.5$ kOe^2. Under our experimental condition ($H \ll H_D$ and $H_\Delta^2 \ll H_0 H_D$) we have

$$\hbar sk/\varepsilon_{1k} = (1 - H/H_0)^{1/2}:$$

The form of the experimental curve $\Delta\omega_k(H)$ thus indicates that $\Delta\omega_k$ is proportional to sk/ε_{1k}.

Figure 4 shows how the parametric magnon relaxation parameter $\Delta\omega_k$ depends on the orientation of the fields **h** and **H** (**h**||**H**) in the base plane of the crystal. The observed 60-degree anisotropy in $\Delta\omega_k$ agrees with the symmetry of the crystal.

DISCUSSION

In the Introduction we have expressions for the relaxation parameters corresponding to the principal intrinsic relaxation processes. Our experimental results for $\Delta\omega_k(k, T, \varepsilon_{1k})$ agree only with Eq. (9), which gives the contribution from three-particle magnon-phonon scattering to the relaxation.

Nonintrinsic relaxation processes may also contribute to magnon relaxation in $FeBO_3$. In particular, $\Delta\omega_k(T)$ has a peak which we attributed in Ref. 8 to a "slow" relaxation channel involving impurity ions. However, the relaxation parameters for the nonintrinsic processes differ from the experimental dependences $\Delta\omega_k(k, T, \varepsilon_{1k})$. In particular,

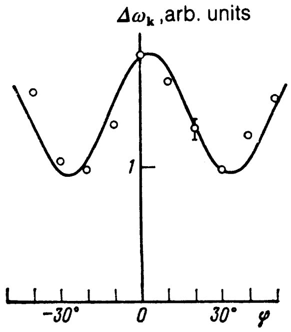

FIG. 4. Dependence of the relaxation parameter $\Delta\omega_k$ on the direction of the fields **h**, and **H** in the base plane of the crystal.

$\Delta\omega_k \propto k$ for elastic scattering processes but is independent of T. According to the data in Ref. 8, the 1–4 K temperature interval of interest lies at the edge of the peak in $\Delta\omega_k(T)$. It is therefore necessary to analyze the inelastic processes (in particular, slow relaxation) with care. The rate of the inelastic scattering process is independent of the magnon wave vector **k** but may depend on the external magnetic field if paramagnetic impurities are involved in the scattering. The interaction energy for the magnetic moment of an impurity ion interacting with a field H ($H < 500$ Oe in our experiment) is much less than the exhange interaction energy for an impurity interacting with its neighbors (the "freezing" temperature of the impurity is $T_i \approx 5$ K, see below). Inelastic processes should therefore not give rise to a strong field-dependence $\Delta\omega_k(H)$. In addition, the experimental finding that $\Delta\omega_k$ was proportional to k at both of the frequencies ω_k (i.e., for fields in two different frequency ranges) further indicates that inelastic scattering processes, and in particular slow relaxation, do not contribute significantly to $\Delta\omega_k$.

We thus conclude that for $1.2 \leqslant T \leqslant 4.2$ K, the time-independent component of the magnon relaxation in $FeBO_3$ is due primarily to two processes, in which a parametric magnon coalesces with a thermal phonon to produce another magnon, or a parametric magnon decays into a magnon and a phonon. This conclusion agrees with the findings in Refs. 21–23, 17, where the properties of a steady-state parametrically excited spin system in $FeBO_3$ were studied for above-threshold pumping fields and three-particle magnon-phonon interactions were found to play an important role in the relaxation to thermodynamic equilibrium.

A magnon-phonon contribution was also reported in Refs. 24 and 25 for the easy-plane antiferromagnets $CsMnCl_3$ and $RbMnCl_3$. However, the procedure used there to identify the magnon-phonon contribution is suspect, because it relies on a theoretical formula in Ref. 15 which does not allow for the dependence of $\Delta\omega_{mp}$ on the magnon wave vector **k**. Our results are thus the first to shed light on the behavior of the magnon-phonon relaxation as a function of the parameters.

The data can be used to find the characteristic magnetoelastic interaction energy Θ for $FeBO_3$. Equation (6) gives $\tau = 1.4 \cdot 10^{-14}$ erg. If (6) is replaced by the expression given in Ref. 13, we obtain $\Theta = 1/2|B_{11} - B_{12}|v_0 = 1.1 \cdot 10^{-14}$ erg, which corresponds to $|B_{11} - B_{12}| = 2.4 \cdot 10^8$ erg/cm^3. These energies Θ are substantially greater than the value Θ_s deduced experimentally from the dependence of the speed of sound on the magnetic field in Refs. 11 and 26, where the values $|B_{14}| = 1.4 \cdot 10^7$ (Ref. 11) and $1.6 \cdot 10^7$ erg/cm^3 (Ref. 26) were found at 77 and 4.2 K, respectively, and the result $|B_{11} - B_{12}| = 2.4 \cdot 10^7$ erg/cm^3 at $T = 77$ K (Ref. 11) was obtained with a large error. Moreover, the magnon-phonon relaxation parameter $\Delta\omega_{mp}$, extrapolated to high temperatures ($T \gtrsim 50$ K) by Eq. (6) with $\Theta \approx 10^{-14}$ erg, is much larger than the total magnon relaxation rate measured previously in Ref. 8 in this temperature range.

This discrepancy can be resolved if the strength of the magnon-phonon interaction is assumed to increase sharply as the temperature decreases (in particular, for $T \lesssim 4$ K). Such an abnormal dependence of Θ on temperature for $T \ll T_N$ could in turn result from switching on an additional magnon-phonon interaction mechanism involving magnetic impurity ions. Since the amplitude for this process depends on the magnetization M_{imp} of the impurity, an abrupt change in Θ could be caused by a large change in M_{imp} in a narrow temperature interval. We note that (as was pointed out by Lutovinov) this additional mechanism could affect Θ and Θ_s differently. The results in Refs. 27 and 28 indicate that $FeBO_3$ may indeed contain impurities whose magnetization changes abruptly for $T \approx 5$–7 K.

Antiferromagnetic resonance in $FeBO_3$ was studied for a wide range of temperatures in Ref. 27, where it was found that the hexagonal anisotropy constant in the base plane of the crystal changes sign at $T_c \approx 5$ K. Hysteresis and broadening of the AFMR line were also observed for $T < T_c$ and were attributed to the presence of impurities. The role of impurities in these effects was subsequently confirmed by theoretical calculation in Ref. 28.

We carried out the following additional experiments to check the validity of the above hypotheses for crystals in which magnon relaxation is observed.

1. We studied the temperature dependence $\Theta_s(T)$ of the magnetoelastic energy in detail. This was done by using the method described in Ref. 26 to analyze the frequency (~ 6 MHz) of the corresponding mode of the elastic oscillations of the crystal. The measurements showed that Θ_s is constant to within 3% for $1.2 \leqslant T \leqslant 77$ K.

2. Since we anticipated that the frequency-doubling efficiency at antiferromagnetic resonance should depend on the magnetic anisotropy and the associated equilibrium alignment of the magnetic moments of the sublattices, we studied the temperature dependence $I_{2\omega}(T)$ of the second-harmonic signal emitted by the crystal. A microwave signal of frequency $\omega/2\pi = 35.4$ GHz was incident on the $FeBO_3$ single crystal while a magnetic field was applied in the base plane. At resonance ($H = H_{AFMR}$) we observed a narrow line of width $\Delta H \approx 10$ Oe whose intensity $I_{2\omega}$ was proportional to the power of the incident signal. We found that the signal $I_{2\omega}$, had a hexagonal anisotropy and changed abruptly (Fig. 5) in the same temperature intervals 5–7 K for which a spin flip transition was noted in Ref. 27. This strongly suggests that our crystals contained the same magnetic impurity as the ones investigated in Ref. 27.

3. The following experiment confirmed that impurities were responsible for the hexagonal anisotropy in the base plane at low temperatures. Magnons are known to interact strongly with magnetic impurities in antiferromagnets.[8] By exciting sufficiently many magnons in the crystal, it should therefore be possible to populate the excited impurity levels and markedly alter the magnetization of the impurity. This should change the anisotropy field when the latter is due to the impurity, and the AFMR line will be shifted.

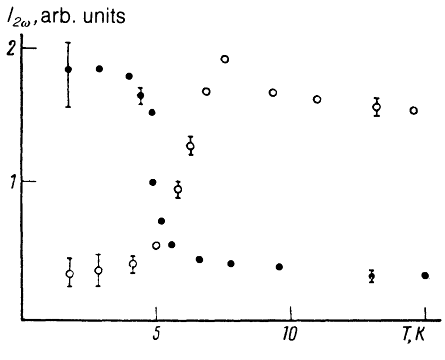

FIG. 5. Temperature dependence of the second-harmonic intensity $I_{2\omega}$ for two orientations φ differing by 30°, with $H = H_{AFMR}$.

In the experiment we placed a crystal inside a two-mode resonator with mode frequencies $\omega_1/2\pi = 35.6$ GHz and $\omega_2/2\pi = 17.5$ GHz. The magnons were excited parametrically by a sequence of rectangular pulses of frequency ω_1, while a continuous signal at the frequency ω_2 was used to observe the AFMR line in the radiation transmitted through the resonator.

The transmitted signal at ω_2 changed by $\sim 10\%$ when a high-power pulse of frequency ω_1 was incident on the crystal. A similar change in the transmitted signal can be achieved by warming the crystal by ≈ 1K. The ω_2 signal relaxed back to its initial value in ~ 20–30 μs. Since this behavior was unchanged when the duration and repetition rate of the ω_1 pulses were varied from 3 to 300 μs and from 25 to 100 Hz, we conclude that it was not caused by simple heating of the crystal. We note that this behavior was observed only for $T < T_c$.

Finally, we point out that the abrupt increase in Θ at low temperatures could be responsible for the peak in $\Delta\omega_{mp}(T)$. In addition to the slow relaxation mechanism, the magnon-phonon interaction may thus also contribute to the peak in $\Delta\omega_{\mathbf{k}}(T)$ $T \approx 18$ K. Indeed, the dependence $\Delta\omega_{\mathbf{k}}(H)$ shown in Fig. 3 for $T = 18$ K is seen to contain a large ($\approx 60\%$) component proportional to the wave vector **k**, indicating a significant magnon-phonon contribution.

CONCLUSION

Our studies show that magnon-phonon interaction is primarily responsible for magnon relaxation in $FeBO_3$ crystals for $T < 4$ K. This interaction is enhanced at low temperatures by magnetic impurity ions in the crystal. The relaxation time for the excited impurity ions is ~ 20–30 μs.

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