Relaxation of magnetoelastic waves in antiferromagnetic $FeBO_3$

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Relaxation of magnetoelastic waves in the easy-plane antiferromagnet $FeBO_3$ is investigated in the frequency range 5–20 MHz. The experiments were performed in the temperature interval 1.5–4.2 K. A peak is observed on the temperature dependence of the relaxation time $\tau_{\rm ph}$ of the magnetoelastic waves, and is apparently due to the presence of Fe^{2+} impurity ions in the crystal. The activation energy of the impurity level $E \approx 30$ K, the components $c_{44} = 1.0 \times 10^7$ erg $\pm$ 10% of the elastic tensor and $B_{14} = 1.6 \times 10^7$ erg/cm^3 $\pm$ 10% of the magnetoelastic tensor, as well as the gap $H_\Delta^2 = 4.9$ kOe^2 $\pm$ 10% in the magnon spectrum are measured at $T = 4.2$ K.

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The study of the relaxation of magnons and phonons in magnetically ordered crystals has shown (see, e.g., Refs. 1 and 2) that its value is strongly influenced by the presence of magnetic impurities in the crystal even at relatively low densities ($\sim$0.01 mol.%). One of the considered mechanism of impurity relaxation of magnons is the so-called slow relaxation that arises when the crystal has local impurity levels with activation energy and with relaxation time $\tau_{\rm imp}$ such that the condition τ^{-1}, $\omega_m(\mathbf{k}) \ll E/\hbar$, is satisfied, where ω_m is the frequency of a magnon with wave vector $\mathbf{k}$. In this case the oscillations of the magnetic moments in the spin wave lead to modulation of the impurity levels, and consequently to their population, with frequency ω_m. Since $\tau_{\rm imp}$ is finite, the change of the population lags the change of the level positions, and the spin-wave energy is dissipated into the lattice via the impurity system. According to the theory expounded in Ref. 1, the relaxation determined by this process is a maximum in the case when

$$\omega_m \tau_{imp} \sim 1. \tag{1}$$

Owing to the magnetoelastic coupling, this mechanism can lead also to relaxation of the phonons. The contribution of the slow relaxation leads to the appearance of characteristic peaks on the temperature dependences of the magnon and phonon relaxation times τ_m and $\tau_{\rm ph}$; the positions T_c of these peaks are determined in accordance with (1) by the $\tau_{\rm imp}(T)$ dependence. This dependence usually obeys the Arrhenius law

$$\tau_{imp} = \tau_0 \exp\,(E/k_B T). \tag{2}$$

Such peaks were noted, in particular, in Refs. 3 and 4, where the line width of the ferromagnetic resonance ($\omega_m(0)/2\pi = 34.5$ GHz, $T_c = 70$ K) and the relaxation of the parametrically excited magnons ($\omega_m(\mathbf{k})/2\pi = 17.7$ GHz, $T_c = 18$ K) were investigated in antiferromagnetic $FeBO_3$ with the magnetic ion Fe^{3+}. The results were interpreted as a manifestation of slow relaxation on impurity centers, due to the presence of Fe^{2+} impurity ions in the investigated crystals.

In Ref. 5, using the samples investigated earlier in Ref. 4, a study was made of the parametric excitation of acoustic phonons of frequency $\omega_{\rm ph}(\mathbf{k})/2\pi = 17.7$ GHz. It follows from the result of this study that the temperature dependences of the relaxation time $\tau_{\rm ph}(T)$ of the parametric phonons also exhibit a peak at $T_c = 18$ K. It was suggested in Ref. 5, on the basis of the equality of the frequencies of the investigated oscillations and of the positions of their relaxation peaks in Refs. 4 and 5, that, first, the observed phonon-relaxation peak is of impurity (Fe^{2+}) origin, and second, that it can be explained by the slow-relaxation mechanism described above.

The present paper is devoted to further study of the nature of the influence of magnetic impurities on the relaxation of the magnons and phonons. To this end we investigated the relaxation of megahertz magnetoelastic oscillations in $FeBO_3$. The magnetic and elastic properties of the easy-plane antiferromagnet $FeBO_3$ ($T_N = 348$ K) have been sufficiently well studied. A summary of the main results and a survey of the literature are contained in Ref. 4. An important factor is the presence of appreciable magnetostriction in $FeBO_3$. The $FeBO_3$ structure is described by the space group D_{3d}^6.

Magnetoelastic interaction in magnets leads to distortion of the unperturbed magnon $\omega_{m_0}(\mathbf{k})$ and phonon $\omega_{{\rm ph}_0}(\mathbf{k})$ spectra; this distortion is particularly strong near their intersection points. The K space region where this perturbation is noticeable is called the overlap region of the spectra. In this region one must speak of the spectrum and properties of normal magnetoelastic oscillations. The spectrum of such oscillations can be obtained in the usual manner, by starting from the thermodynamic potential $\Phi = \Phi_{\rm m} + \Phi_{\rm el} + \Phi_{\rm me}$ which takes into account the magnetic, elastic, and magnetoelastic interactions. Concrete expressions for each of these terms, which are determined by the crystal symmetry, are given in Refs. 6–8. The spectra of the magnons and phonons, with account of their interaction (magnetoelastic waves), depend strongly on the ratio of the limiting (in the limit of large k, but when the continuous-medium approximation is still applicable, $k \ll \pi/a_0$, where a_0 is the size of the unit cell) magnon propagation velocity s and the speed of sound c. In $FeBO_3$, $s > c$ for all propagation directions. In this case the unperturbed spectra of the magnons and phonons do not intersect and it is possible to subdivide arbitrarily the elementary excitations corresponding to the magnetoelastic

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waves into quasiphonons and quasimagnons, in accord with their dispersion laws outside the spectrum-overlap region.

We shall be interested hereafter in transversely polarized quasiphonons propagating along the principal axis C_3 and whose spectrum distortion due to the magnetoelastic interaction is a maximum. In the case when the magnetic field **H** is in the basal plane of the crystal ($z\|C_3$, $x\|\mathbf{H}$), these are quasiphonons polarized along **H**. According to Refs. 9 and 10 their spectrum can be represented in the form

$$\omega_{ph}(k)=c_{tz}\left(1-\frac{\gamma^2\lambda^4 l_0^3 H_E/\mu_5}{\omega_m^2}\right)^{1/2}k, \tag{3}$$

where ω_m is the frequency of quasimagnons with wave vector **k** and belonging to the low-frequency of the spectrum, is determined in our case by the expression[8,11,12]

$$\omega_m^2(k)=\gamma^2[H(H+H_D)+H_\Delta^2+\alpha_\|^2k^2], \tag{4}$$

$c_{tz}=c_{44}/\rho)^{1/2}$ is the speed of the corresponding sound in the limit as $H\to\infty$, $\mu_5=c_{44}/2$ 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 sublattice magnetization, H_E is the exchange field, γ is the gyromagnetic ratio, H_D is the Dzyaloshinskiĭ field, H_Δ^2 is a parameter determined mainly by the magnetoelastic interaction, and $\alpha_\|$ is an exchange constant (μ and λ are the symbols adopted in Refs. 7 and 8 for the parameters of the thermodynamic potential). We have neglected in (4) the dipole interaction and the magnetic anisotropy in the basal plane, which are immaterial in our case.

PROCEDURE AND SAMPLES

The magnetoelastic waves were exicted in $FeBO_3$ by a high-frequency magnetic field **h**. A homemade pulsed RF oscillator could be tuned in the interval 5–20 MHz with the aid of a variable capacitor and by changing the number of turns of a removable tank-circuit coil (3–20 turns). The RF-pulse repetition frequency was 50 Hz, the pulse duration 1 msec, and the amplitude falloff time ~ 1 μsec. The amplitude of the RF pulse on the coil was ~ 2 V. The narrow face of the sample was secured with BF-2 adhesive to the end of a platinum rod soldered through the wall of a hermetically sealed glass ampoule. To improve the heat transfer from the sample, the ampoule was filled with helium gas at a pressure ~ 10 Torr. The ampoule with the sample were placed in the coil of the tank circuit of the RF oscillator, and the entire assembly was placed in a solenoid secured in a dewar with liquid helium and producing a constant magnetic field **H**. The fields **h** and **H** were oriented in the basal plane of the crystal and perpendicular to each other. The field H was measured accurate to $\sim 1\%$.

The intensity of the magnetoelastic oscillations in the sample was recorded by measuring the emf induced in the exciting coil after turning on the RF pulse. The time variation of this emf was investigated with the aid of an oscilloscope whose sweep was triggered by the trailing edge of the RF pulse. The oscillation relaxation time τ_{ph} was defined as the time in which the amplitude of the signal decreased by a factor e.

The measurements were made in the temperature interval 1.5–4.2 K. The temperature was determined, by measuring the saturated vapor pressure of helium, at an accuracy not worse than 0.1 K. By varying the repetition rate and the amplitude of the RF pulses it was possible to verify that the overheating of the sample by the RF field did not exceed the temperature-measurement error. We investigated two $FeBO_3$ samples in the form of polished plates with transverse dimensions $\approx 2\times 2$ mm and thickness 0.18 and 0.14 mm. The samples were cut from one single crystal. The planes of the plates coincided with the basal plane of the crystal.

EXPERIMENTAL RESULTS

By varying smoothly the RF generator frequency, we found that at certain frequencies the magnitude and duration of the signal induced in the coil increased resonantly. The amplitude of this signal decreased with time exponentially and was slightly modulated at the low frequency. The indicated resonances correspond to excitation in the sample of different modes of the natural magnetoelastic oscillations. From among these we chose for further study, on the basis of the known sample thickness d and speed of sound c_{tz} at $T=293$ K (see Ref. 9), the mode corresponding to the first harmonic of the resonance of the plane transverse magnetoelastic wave over the crystal thickness ($\mathbf{k}\|C3$, $|\mathbf{k}|=\pi/d$).

Figure 1 shows the $\omega_{ph}(H)$ dependence of this mode for one of the samples, measured at $T=4.2$. From the experimental data and the known values of the parameters in (3) and (4), namely $H_E=2.6\times 10^6$ Oe, $H_D=1.08\times 10^5$ Oe, $\gamma=2\pi\cdot 2.8$ GHz/kOe, $l_0=1.0\times 10^3$ G (the term $\alpha_\| k_\|^2$ in (3) can be neglected in our case), we can determine the values $c_{tz}=4.85\times 10^5$ cm/sec $\pm 2\%$, $\lambda_4/l_0^2=3.2\times 10^7$ erg/cm^3 $\pm 10\%$, $H_\Delta^2=4.9$ kOe^2 $\pm 10\%$. Such measurements were performed by us also at $T=77$ and 293 K. An earlier investigation of the $\omega_{ph}(H)$ dependence for the studied mode of the magnetoelastic oscillations in $FeBO_3$ at $T=77$ and 293 K was carried out in Ref. 9. The experimental data obtained by us at these temperatures agree within the limits of experimental accuracy with those of Ref. 9, but the value of H_E used in the reduction of the experimental data in Ref. 9 was approximately twice as large.

Figure 2 shows the results of the investigation of the temperature dependence of the relaxation time τ_{ph} of the magnetoelastic oscillations at certain values of the magnetic field H. It follows from Fig. 2 that besides the considerable

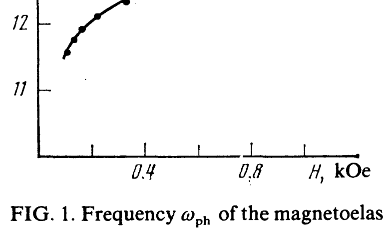

FIG. 1. Frequency ω_{ph} of the magnetoelastic oscillations vs the magnetic field H at $T=4.2$ K and $d=0.18$ mm. Solid line—$\omega_{ph}(H)$ dependence calculated from (5) with the parameters whose values are given in the text.

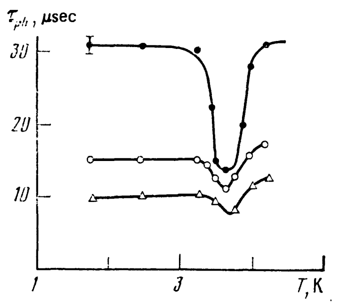

FIG. 2. Temperature dependences of the relaxation time $\tau_{\rm ph}$ at: △—$H = 0.25$, ○—0.45, ●—0.9 kOe ($d = 0.18$ mm).

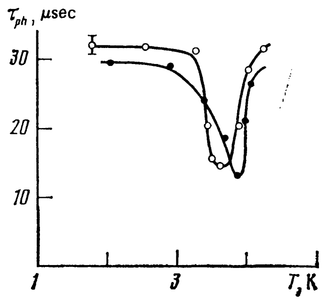

FIG. 3. Temperature dependences of $\tau_{\rm ph}$ at various sample thicknesses: ○—$d = 0.18$, ● $= 0.14$ mm ($H = 0.9$ kOe).

increase of $\tau_{\rm ph}$ with increasing H, a sharp decrease of the relaxation time is observed in the investigated field interval at $T = T_c = 3.6 \pm 0.1$ K. We note also that the relaxation time $\tau_{\rm ph}$ increases with the field more slowly at $T \approx T_c$. The $\tau_{\rm ph}(T)$ dependences measured for the other sample are similar in form.

Figure 3 shows the $\tau_{\rm ph}(T)$ dependences measured at a single value of the field at both samples. It can be seen that for the thinner sample, which corresponds to a higher natural frequency $\omega_{\rm ph}$, the relaxation peak is shifted towards higher temperatures.

DISCUSSION OF RESULTS

Investigations of the equations of motion that describe the spin and elastic oscillation in a magnetoelastic wave make it possible not only to determine its dispersion law, but also to express the magnetoelastic-wave relaxation time in terms of the relaxation times of the interacting spin and elastic waves. We introduce for this purpose the complex frequencies

$$\Omega_m = \omega_m + i\tau_m^{-1}, \quad \Omega_{ph0} = \omega_{ph0} + i\tau_{ph0}^{-1},$$

where τ_m and $\tau_{\rm ph0}$ are respectively the relaxation times of the quasispin and elastic waves. Substituting them in the secular equation that determines the frequency $\Omega_{\rm ph} = \omega_{\rm ph} + i\tau_{\rm ph}^{-1}$ of the magnetoelastic wave, we obtain for the natural frequency and for the relaxation time of the investigated type of magnetoelastic oscillations in a thin plate the following expressions:

$$\omega_{ph} = \omega_{ph0}(1-\delta)^{1/2}, \tag{5}$$

$$\tau_{ph}^{-1} = \frac{\tau_{ph0}^{-1}}{(1-\delta)^{1/2}} + \frac{\omega_{ph0}}{\omega_m}\frac{\delta\tau_m^{-1}}{(1-\delta)^{1/2}}, \tag{6}$$

where

$$\delta = \frac{\lambda_4^2 H_E l_0^3 \gamma^2}{\mu_5 \omega_m^2}, \quad \omega_{ph0} = \frac{\pi c_{tz}}{d},$$

and ω_m is defined by Eq. (4).

The measured $\tau_{\rm ph}(H)$ dependence of both samples is well described by expression (6) with values of $\tau_{\rm ph0}$ and τ_m independent of H. Outside the relaxation peak their value are $\tau_{\rm ph0}^{-1} = 10^5$ sec^{-1} and $\tau_m^{-1} \approx 5 \times 10^5$ sec^{-1}. At $T = T_c$ the parameter τ_p^{-1} is increased by approximately three times, while τ_m^{-1} remains unchanged within the limits of experimental accuracy. The obtained value of τ_m agrees with the homogeneous-magnetization-precession relaxation time determined from the antiferromagnetic-resonance line width. The value of $\tau_{\rm ph0}$ corresponds to an acoustic $Q_{\rm ph} \sim 4 \times 10^3$, approximately five times smaller than the value of $Q_{\rm ph}$ obtained in Ref. 5 for the measured type of oscillations at $k \sim 10^5$ cm^{-1}. It is possible that this discrepancy is due to the additional relaxation on the sample boundaries, a relaxation that depends, in particular, on the sample mounting and is immaterial at large k. Naturally, no relaxation peak was observed on the $\tau_m(T)$ plot, since the relaxation, determined by the impurity mechanism, for which $\Delta\tau_m^{-1}/2\pi \sim 10$ MHz, according to Ref. 4, is much smaller than τ_m^{-1}.

The peaks observed on the $\tau_{\rm ph}(T)$ plot are determined apparently by the slow-relaxation mechanism described above, on impurity levels that are of the same type as those noted earlier in Refs. 3–5. Favoring this assumption are the following facts: first, the temperature T_c at which the relaxation peak is observed increases in accord with (1) and (2) with increasing frequency $\omega_{\rm ph}$; second, for the impurity-level activation energy determined according to Eqs. (1) and (2), the data of the present paper and of Refs. 4 and 5 yield a value $E/k_B \approx 30$ K, which is close to the value $E/k_B \approx 20$ K determined from the data of Refs. 3 and 4. It is also important that the crystals investigated in the present study and in Refs. 4 and 6 were from the same growth batch.

We proceed now to discuss the nature of the impurity levels that lead to the observed peculiarities of the relaxation. It was suggested in Ref. 4 that the slow relaxation observed in FeBO$_3$ is due to $\mathrm{Fe}^{2+} \leftrightarrow \mathrm{Fe}^{3+}$ electron jumps. It should be noted that in the FeBO$_3$ the positions occupied by the Fe ions are crystallographically equivalent,[13] therefore the spin and elastic oscillations should seemingly not cause the indicated jumps. For the crystal to be electrically neutral, however, the presence of Fe^{2+} ions in it is always accompanied by the presence of impurity ions with valence higher than three. These ions M form with the Fe^{2+} ions, at $T \lesssim 100$ K, bound states that lead to violation of the local symmetry at the Fe^{2+} ions. In this case the energy levels corresponding to different positions of the Fe^{2+} ions turn out to depend differently on the spin direction, and the spin wave induces, as indicated in Ref, 14, the $\mathrm{Fe}^{2+} \leftrightarrow \mathrm{Fe}^{3+}$ electron jumps. The Fe^{2+} ions have a relatively strong spin-orbit interaction, therfore the modulation of the crystal field at the Fe^{2+} ion, due to the elastic wave, should also lead to electron jumps and accordingly to relaxation of the elastic wave.

The activation energy E determined by us is close to those measured for hematite[14] and yttrium iron garnet[15] crystals. In these references the impurity relaxation was also attributed to the jump-over mechanism. Such a relatively low value of E can be attributed to the smallness of the splitting, induced by the neighboring ion M, of the levels of Fe^{2+}.

Unfortunately, the experiments performed do not exclude the possibility that the slow relaxation is due to intra-ion transitions in the Fe^{2+} impurity ions. To cast light on this problem it would be useful to investigate crystals with different impurity contents.

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