Study of parametric excitation of magnons and phonons in antiferromagnetic $FeBO_3$

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Parametric excitation of magnons and phonons in the easy-plane antiferromagnet $FeBO_3$ is investigated by microwave pumping at a frequency $\nu_p = 35.4$ GHz. The experiments are performed at temperatures from 1.2 to 180 K. The dependences of the phonon relaxation frequency on the temperature and on the magnetic field strength are measured. They indicate that phonon-magnon interaction contributes significantly to the phonon relaxation. At $T = 18$ K the phonon relaxation exhibits a maximum that can be attributed to the presence of Fe^{++} impurity ions in the crystal.

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Starting with the studies of Bloembergen and Damon[1] and of Suhl,[2] the method of parametric excitation of inhomogeneous oscillations is being successfully used to investigate the properties of elementary excitation of the spin systems of magnetically ordered substances. Parametric excitation of magnons is made possible by the always present nonlinearity of the equation of motion of the magnetic moments, although the nature of this nonlinearity in magnets having different magnetic structures may vary.[3,4]

Magnetoelastic interaction in magnets leads to disturtion of the unperturbed magnon and phonon spectra, $\omega^0_{im}(\mathbf{k})$ and $\omega^0_{iph}(\mathbf{k})$, respectively, a distortion particularly strong near the their intersection point, as well as to a change of the damping of the magnons and phonons.[5,6] It must be noted here that in the general case one should speak of the spectrum and properties of normal magnetoelastic oscillations, but outside the so-called spectrum-overlap region the concepts "magnon" and "phonon" remain perfectly adequate. Fulfillment of this condition is implied throughout.

Magnetoelastic interaction makes possible also parametric excitation of oscillations of not only the magnetic system but also of the elastic system by a high-frequency magnetic field. These processes were first investigated in ferrites (see, e.g., Refs. 7 and 8). A detailed review of the results of these researches are given in the books of Le Crow and Comstock[9] and of Tucker and Rampton.[10]

The phenomena described should be particularly strongly pronounced in antiferromagnets with anisotropy of the easy-plane type, a characteristic feature of which is exchange enhancement of the magnetoelastic interaction and the presence of a low-activation mode of the magnon spectrum.[11–13]

In the language of quasiparticles, the elementary act of parametric excitation reduces to absorption, by the sample, of one or several photons (p) with simultaneous production of two or more elementary excitations corresponding to oscillations of the magnetic (m) and elastic (ph) systems. The number of secondary excitations increases exponentially when the high-frequency magnetic field $|\mathbf{h}|$ at the sample exceeds a certain threshold value h_c. The lowest threshold is usually possessed by a three-particle process for which the conservation laws are of the form

$$\omega_p=\omega_\alpha(\mathbf{k}_1)+\omega_\beta(\mathbf{k}_2), \quad \mathbf{k}_1+\mathbf{k}_2=\mathbf{k}_p\approx 0, \tag{1}$$

where α, $\beta = m, ph$. Parametric excitation is produced in experiment by perpendicular and parallel pumping, so designated in accord with the relative orientation of the high-frequency and static fields $\mathbf{h}$ and $\mathbf{H}$, respectively.

Depending on the excitation method, on the pump frequency, and on the parameters that determine the elementary excitation spectra and relaxation frequencies, particle pairs belonging to different branches of the magnon and phonon spectra can be excited. Excitations of magnon ($p \to m + m$),[14,15] mixed ($p \to m + ph$),[16] and phonon ($p \to ph + ph$) pairs[17,18] in easy-plane antiferromagnets have been investigated.

This paper is devoted to the study of parametric instabilities in the easy-plane antiferromagnet $FeBO_3$. This is one of the few known antiferromagnets with the combination of high Néel temperature ($T_N = 348$ K), appreciable magnetostriction, strong Dzyaloshinskiĭ interaction, and optical transparency. The magnetic and elastic properties of $FeBO_3$ were investigated by various methods. A summary of the main results and a review of the literature are given in Ref. 19.

The structure of $FeBO_3$ is described by the space group D^6_{3d}. The thermodynamic potential with allowance for the magnetoelastic interaction can be represented in the form

$$\Phi=\Phi_m+\Phi_{me}+\Phi_{el},$$

where Φ_m, Φ_{me}, and Φ_{el} are respectively the magnetic, magnetoelastic, and elastic terms. Concrete expressions determined by the crystal symmetry are given for each term of Φ in Refs. 20, 12, and 21.

The spectra of the magnons and phonons, with account taken of their interaction, depend significantly on the ratio of the maximum magnon propagation velocity s and the

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sound velocity v. In $FeBO_3$ we have $s > c$. The magnon spectrum of a two-sublattice antiferromagnet consists of two branches, which are described in the case of $FeBO_3$, in the continuous-medium approximation ($d^{-1} \ll |\mathbf{k}| \ll \pi/a_0$, where d is the dimension of the sample and a_0 is the lattice constant) and when $\mathbf{H}$ lies in the basal plane of the crystal, by the following equations[11,12,22]:

$$(\omega_{1km}/\gamma)^2 = H(H+H_D) + H_\Delta^2 + 36 H_A$$

$$H_E \cos 6\varphi + \alpha_\parallel^2 k_\parallel^2 + \alpha_\perp^2 k_\perp^2 + H_{\text{dip}}^2, \qquad (2)$$

$$(\omega_{2km}/\gamma)^2 = 2H_A H_E + H_D(H+H_D) + \alpha_\parallel^2 k_\parallel^2 + \alpha_\perp^2 k_\perp^2, \qquad (3)$$

where ω_{ikm} and $\mathbf{k}$ are the cyclic frequency and the wave vector of the magnon, $\gamma = 2\pi \times 2.8$ GHz/kOe is the gyromagnetic ratio, H_E is the exchange field, H_D is the Dzyaloshinskiĭ field, H_A is the uniaxial anisotropy field, $H_A^{(6)}$ is the hexagonal anisotropy field in the basal plane, H_Δ^2 is a spectrum parameter governed mainly by the magnetoelastic interaction, $\alpha_\parallel$ and $\alpha_\perp$ are exchange constants (the subscripts $\parallel$ and $\perp$ indicate the direction of $\mathbf{k}$ relative to the principal axis C_3), and H_{dip}^2 is a parameter governed by the dipole-dipole interaction. In an orthogonal coordinate frame with $z \parallel C_3$ and $x \parallel \mathbf{H}$, with isotropy of the exchange constants ($\alpha_\parallel = \alpha_\perp = \alpha$) and $4\pi\chi_\perp \ll 1$ ($\chi_\perp = M_0/H_E$ is the perpendicular susceptibility) we have according to Ref. 23

$$H_{\text{dip}}^2 = 4\pi\chi_\perp [H(H+H_D) + \alpha^2 k^2] \left[\frac{k_z^2}{k^2} + \frac{(H+H_D)^2}{H(H+H_D)+\alpha^2 k^2} \frac{k_y^2}{k^2}\right]. \qquad (4)$$

According to Ref. 24, the spectrum of the transversely polarized phonons, which are usually more strongly coupled with the magnons, is of the form

$$\omega_{ph} = c\left(1 - \frac{4\gamma^2 H_E B_{15}/M_0 c_{44}}{\omega_{1km}^2}\right)^{1/2} k, \qquad (5)$$

where c is the speed of the corresponding sound in the limit as $H \to \infty$, B_{15} is a component of the magnetostriction tensor, c_{44} is the elastic modulus, and M_0 is the sublattice magnetization.

PROCEDURE AND SAMPLES

The parametric excitation of magnons and phonons was investigated with a direct-amplification spectrometer by a procedure described in detail in Ref. 25. We used a high-Q cylindrical cavity ($Q \approx 10^4$) tuned to the H_{012} mode. The sample was fastened to the bottom of the cavity in an antinode of the microwave magnetic field $\mathbf{h}$ in a small container made of cigarette paper to eliminate the elastic stresses that are produced when the sample is cooled and influence noticeably the magnon spectrum in $FeBO_3$. The fields $\mathbf{h}$ and $\mathbf{H}$ were in the basal plane of the crystal, and the direction of $\mathbf{H}$ in this plane could change in the course of the experiment.

Long pulses from a cw magnetron microwave oscillator operating at 35.4 GHz and 10 W were obtained by modulating the oscillator-anode voltage. The pulse duration could be varied from 0.01 to 1 msec. To weaken the power-supply interference, the 50-Hz supply was used to trigger the oscillator.

The measurements were made in a temperature range 1.2–180 K. To improve heat transfer from the sample the cavity with the sample were filled with liquid helium at $T \leqslant 4.2$ K or were cooled with helium vapor at higher temperatures. The temperature was measured with a semiconductor resistance thermometer. The accuracy with which the temperature was measured and maintained was not worse than 2%.

The microwave pulse passing through the cavity with the sample was detected and displayed on an oscilloscope screen. The power P extracted from the resonator through a weakly coupled opening, proportional to the squared microwave field h^2 at the sample, was measured with a square-law detector calibrated with a thermistor power meter. The field h at the sample was determined from the value of P and from the cavity parameters with absolute accuracy 20%, but the relative accuracy was much better, $\sim 3\%$.

The single-crystal $FeBO_3$ samples were naturally faceted plates measuring $2 \times 2 \times 0.5$ mm. The plate growth plane coincided with the basal plane of the crystal.

MEASUREMENT RESULTS

The experiments have shown that starting with a certain threshold level of the microwave field h at the sample in weak static fields, the pulse passing through the resonator undergoes a characteristic distortion. The form of the distortion indicates that absorption of the microwave power by the sample develops in the course of time. Thus, the picture usually observed in parameteric excitation was obtained. Figure 1 shows the results of an investigation of the dependence of the threshold field h_c on the direction of $\mathbf{H}$ in the basal plane (φ is the angle between the vectors $\mathbf{h}$ and $\mathbf{H}$) at $H = 330$ Oe. It follows from the form of the $h_c(\varphi)$ dependence and from the crystal symmetry that h_c is indeed determined by the angle φ between the fields $\mathbf{h}$ and $\mathbf{H}$ and not by the direction of $\mathbf{H}$ relative to the crystallographic axes of the crystal. It follows also from Fig. 1 that two different parametric processes are observed, parallel ($h_{c\parallel}$) orientation of the fields $\mathbf{h}$ and $\mathbf{H}$ being optimal for one of them, and perpendicular ($h_{c\perp}$) for the other. These processes have different values of h_c at the orientation of $\mathbf{h}$ and $\mathbf{H}$ that is optimal for a given process, different instability-development times, and different field and temperature dependences of the threshold fields. It is important also that both processes can be observed at certain values of the angle φ.

The solid curves in Fig. 1 are plots of $h_{c\parallel}/|\cos\varphi|$ and $h_{c\perp}/\cos(\varphi - \pi/2)$. From the fact that the experimental points fit these plots with good accuracy it follows that the indicated parametric processes are excited by the corresponding projections of the microwave field $\mathbf{h}$ on the static $\mathbf{H}$.

The instability excited by the parallel component of $\mathbf{h}$ (parallel pumping) was investigated in detail by us in Ref. 19. It was shown there that parallel pumping causes parametric excitation of magnons with a frequency ω_{1km} equal to half the pump frequency ω_p, and with wave vectors $\mathbf{k}$ given by Eq. (2).

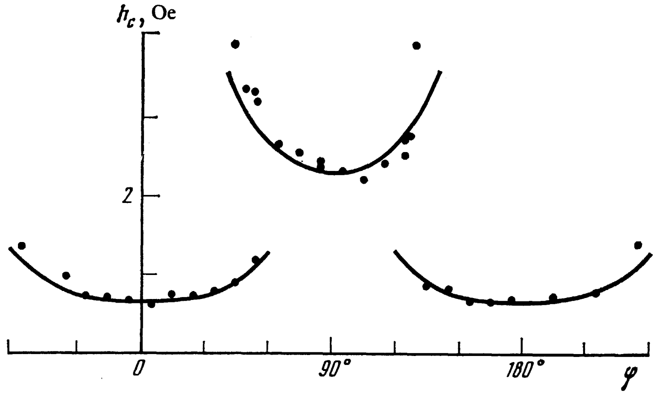

FIG. 1. Dependence of the parametric instability threshold field h_c on the angle φ between the directions of the fields **h** and **H** applied in the basal plane of the crystal. $H = 330$ Oe, $T = 77$ K. The solid lines are plots of $h_{c\perp}/\cos(\varphi - \pi/2)$ and $h_{c\parallel}/|\cos\varphi|$.

We dwell now on the investigation of the instability that develops under perpendicular pumping. The parametric excitation at $\mathbf{h} \perp \mathbf{H}$ proceeds as follows. The microwave pump field **h** excites at a frequency ω_p uniform magnetic-moment oscillations corresponding to the low-activation branch of the magnon spectrum. When a certain critical amplitude of the oscillations is reached, buildup of a pair of inhomogeneous oscillations becomes possible, with wave vectors $\mathbf{k}_i$, in both the magnetic (m) and elastic (ph) systems at frequencies ($\omega_\alpha(\mathbf{k}_i)$ ($\alpha = m, ph; i = 1,2$). As already indicated, the lowest threshold is possessed by the "first-order Suhl instability," for which relations (1) hold.

Figure 2 shows the dependence of the threshold field of this instability, h_c, on the static field H at $T = 90$ K. The plot of $h_c(H)$ is similar also for other temperature. The fact that h_c decreases when H approaches H_R (the field corresponding to antiferromagnetic resonance (AFMR) at the frequency ω_p) and that the instability is excited by perpendicular pumping means that we are observing a Suhl instability. And from the fact that the parametric instability is observed at both $H < H_R$ and $H > H_R$ it follows apparently that the excited particles are phonons, inasmuch is in accordance with (2) the magnon frequency is higher than ω_p at $H > H_R$.

Figure 3 shows plots of $h_c(T)$ at $H < H_R$ and $H > H_R$. It can be seen from Fig. 3 that both plots have peaks at $T \sim 18$ K. A peak is observed at this temperature in the entire investigated range of static magnetic fields H.

DISCUSSION OF RESULTS

Parametric excitation of phonons in antiferromagnets with easy-plane anisotropy at first-order Suhl instability was considered theoretically by Lutovinov and Savchenko (Ref. 26).[1] They obtained the following expression for the threshold field of the process:

$$h_c = \xi\left(\frac{\hbar}{\gamma}\right)^2 \frac{\omega_0^2+(\omega_p/2)^2[(s/c)^2-1]}{\theta^2}\,\frac{\omega_0}{H+H_D}\,\frac{Mc^2}{J_0} \times \frac{(\omega_p^2-\omega_0^2)^2+\Delta\omega_0^2[(\omega_p-\omega_0)^2+(\omega_p+\omega_0)^2]}{2\omega_p[\omega_0^2(\omega_p^2-\omega_0^2)^2+\Delta\omega_0^2(\omega_p^2+\omega_0^2)^2]^{1/2}}(\eta_1\eta_2)^{1/2}, \tag{6}$$

where $\xi \sim 1$ is a parameter governed by the anisotropies of the magnetoelastic interaction and of the phonon relaxation, $\hbar$ is Planck's constant, ω_0 is the AFMR frequency determined by Eq. (2) at $\mathbf{k} = 0$, $\theta = B_i V_0 \sim 2\times 10^{-15}$ erg (Ref. 27)[2] is the characteristic energy of the magnetoelastic interaction, B_i is a linear combination of the corresponding components of the magnetostriction tensor, V_0 and M are respectively the volume and mass of the unit cell, $s = \alpha\gamma$ and c are the maximum propagation velocities of the magnons and phonons, J_0 is the exchange integral, $\Delta\omega_0$ is the AFMR line width, and η_1 and η_2 are the relaxation parameters of the excited phonons (the phonon lifetime is $\pi = \eta^{-1}$).

The $FeBO_3$ parameters in Eq. (6) were taken from Ref. 19. Some of them, e.g., the magnetoelastic constants, were measured not in the entire investigated temperature interval,

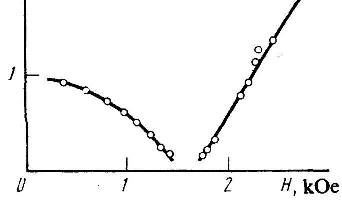

FIG. 2. Dependence of the threshold field h_c on the static field H, $T = 90$ K.

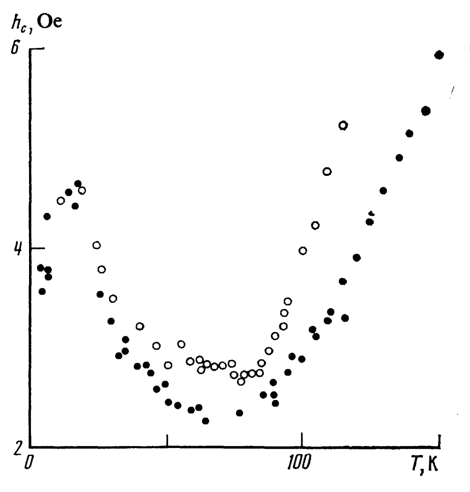

FIG. 3. Plots of $h_c(T)$ at $H < H_R$ and $H > H_R$; ●—$H = 330$ Oe, ○—2.34 kOe.

but can be easily extrapolated from the known function $M_0(T)$, which is proportional to Brillouin function $B_{5/2}(T)$ ($B_i \propto M_0^2$, $J_0 \propto H_D \propto s \propto M_0$). We note *in passim* that owing to the high Néel temperature M_0 changes insignificantly at low temperatures, by 2% when T is changed from 1.2 to 77 K. Equation (6) was obtained under the assumption that the AFMR line has a Lorentz shape. Assuming that a first-order instability has been observed in our experiment and that phonon pairs belonging to one branch of the spectrum are excited, with wave vectors **k** and $-$**k** [see (1)] and $\omega_{ph}(\mathbf{k}) = \omega_p/2$, we put $\eta_1 = \eta_2 = \eta$. This assumption agrees with the results of Ref. 18, in which parametric excitation of phonons of frequency $\omega_{ph}(\mathbf{k}) = \omega_p/2$, propagating along the C_3 axis was observed in $FeBO_3$ at room temperature and under perpendicular pumping at a frequency $\omega_p/2\pi = 9$ GHz by the Mandel'shtam-Brillouin light scattering method.

Using Eq. (6) we can calculate the phonon relaxation parameter η from the experimental values of h_c. An important factor in the determination of η, in our opinion, is that we have succeeded in observing parametric excitation of phonons in a wide range of fields **H**. The point is that in all the Suhl-instability investigations known to us were made either in a resonant or nearly resonant field. Therefore when an attempt is made to determine the relaxation of the excited particles from the threshold field h_c of the process the true resonant-line width $\Delta\omega_0$ contained in the $h_c(\eta)$ dependence was under question. Far from field resonance, when $\omega_p^2 - \omega_0^2 \gg \Delta\omega_0^2$, we can set $\Delta\omega_0$ equal to zero in Eq. (6).

The question remains, to be sure, whether the AFMR line is the Lorentzian $\chi(\omega)$ assumed in the derivation of (6). The $\chi(\omega)$ dependence was theoretically investigated for ferromagnets in Ref. 28. We know of no such studies for antiferromagnets, and since the relative frequency change $\delta\omega_0/\omega_p$ is not large in our experiment, this approximation seems acceptable to us.

The phonon relaxation thus calculated as a function of the temperature and of the magnetic field is shown in Figs. 4

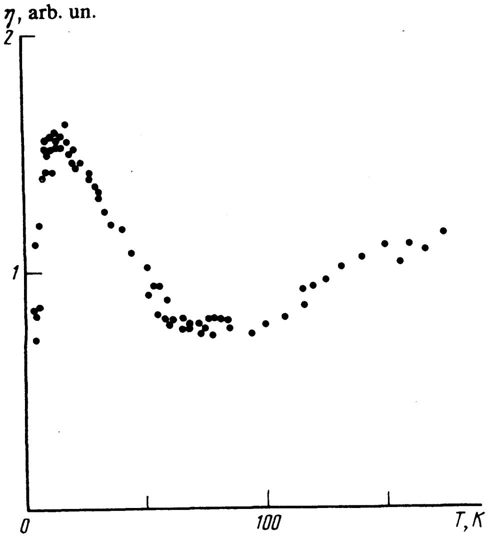

FIG. 4. Temperature dependence of the phonon relaxation parameter η at $H = 1.21$ kOe.

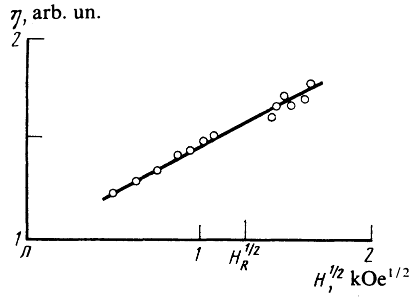

FIG. 5. Phonon relaxation parameter vs magnetic field at $T = 90$ K.

and 5. The relaxation is given in these figures in arbitrary units, since Eq. (6) contains the parameters ξ and θ, which are known only in order of magnitude. An estimate for $H \to 0$ and $T = 90$ K yields a value $\eta/2\pi \approx 1$ MHz, corresponding to an acoustic quality factor $Q_{ph} = \omega_{ph}/2 \approx 2 \times 10^4$.

Figure 5 does not show data corresponding to the field interval $|H - H_R| < \Delta H_0|2 = 60$ Oe, where ΔH_0 is the AFMR line width, since their use, as indicated above, calls for knowledge of the exact AFMR line shape.

We proceed now to discuss the nature of the phonon relaxation in $FeBO_3$. A fairly complete review of the theoretical studies of hypersound damping in dielectrics is contained in the monograph by Tucker and Rampton.[10] An important parameter in the theory is the product of the phonon frequency ω_{ph} by the average lifetime τ of the thermal phonons in the crystal. There are two greatly differing limiting cases: $\omega_{ph}\tau \gg 1$ and $\omega_{ph}\tau \ll 1$. In the first case, which was considered by Landau and Rumer,[29] the relation

$$\eta \propto \omega_{ph} T^4 \tag{7}$$

was obtained. In dielectrics this equation holds for $\omega_{ph}/2\pi \sim 10$ GHz below 50 K.

In the second case, investigated by Akhiezer[30] (see also Ref. 10, Chap. 4), relations

$$\eta \propto \omega_{ph}^2/T \tag{8}$$

were obtained at temperatures low compared with the Debye temperature T_D, and

$$\eta \propto \varkappa T \omega_{ph}^2 \tag{9}$$

at high temperatures. As shown by Akhiezer, at $T \gg T_D$ the thermal conductivity is $\varkappa \propto T^{-1}$, and in this case η is independent of temperature. From the known sound velocities we can estimate the Debye temperature in $FeBO_3$ at $T_D \approx 200$ K.

In magnetic dielectrics it is possible also to have phonon relaxation with participation of magnons. These processes in antiferromagnets with easy-plane anisotropy were considered by Lutovinov, Preobrazhenskiĭ, and Semin.[6] According to estimate based on their results the contribution of these processes should be negligible in our case.

As seen from Fig. 4, at low temperatures a peak of $\eta(T)$ is observed at $T \sim 18$ K, rather than the theoretically predicted monotonic relation. A similar anomaly was observed by us at the same temperature for the magnon relaxation in the same crystals.[19] We have attributed this phenomenon to the presence, in the investigated crystals, of impurity levels connected with the presence of Fe^{++} ions in the crystal. When the magnetic moments oscillate with frequency ω these levels are modulated, as are therefore their populations, at the same frequency. This modulation leads to the so-called slow relaxation. According to theory,[31] the maximum damping should be observed at a temperature at which the condition

$$\omega\tau_{imp}=1 \quad (10)$$

is satisfied, where τ_{imp} is the relaxation time of the impurity ion and varies like $\tau_{imp} = \tau_0 \exp(E/k_B T)$.

Owing to the magnetoelastic coupling, this mechanism should lead to phonon relaxation. We assume therefore that the temperature maximum observed in the phonon relaxation is also due to the presence of the Fe^{++} impurity in the crystal. Since the temperatures at which the magnon and phonon dampings are equal, it follows from the condition (10) that in our experiment the frequency of the excited phonons coincides at $\mathbf{h}\perp\mathbf{H}$ with the frequency of the magnons excited at $\mathbf{h}\|\mathbf{H}$, i.e., $\omega_m = \omega_{ph} = \omega_{p/2}$. This confirms our assumption that under perpendicular pumping we observe a "Suhl instability of first order."

Thus, the presence of a relaxation peak due to an impurity prevents a comparison with the theory at low temperature. At higher temperatures, a noticeable increase of the relaxation with temperature is observed. This contradicts the theory[30] that predicts for nonmagnetic dielectrics a decrease of η with increasing T in this region. It follows hence, in our opinion, that the phonon relaxation in $FeBO_3$ at high temperatures is determined mainly phonon-magnon interaction.

Also noteworthy is the considerable growth of the phonon damping with increasing magnetic field (see Fig. 5). The very dependence of η on the magnetic field also points to a substantial contribution of processes in which magnons participate to the phonon relaxation.

The observed $\eta(H)$ dependence, however, cannot be described by any theoretical formulas known to us. As already indicated in Ref. 19, in $FeBO_3$ the intersection of the unperturbed spectra of the magnons and phonons occurs at $H = 0$ and $\mathbf{k} = 0$ (if weak magnetic interactions are neglected). With increasing magnetic field the spectra move apart, and this should apparently weaken the mutual influence of the magnetic and elastic systems. The observed $\eta(H)$ dependence can be explained if it is assumed that the lower oscillations have a higher Q than the magnetic-system oscillations coupled to them.

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1) The authors thank V. S. Lutovinov for reporting the corrected Eq. (6).
2) Owing to a misprint, the value given for θ in Ref. 19 is larger by two orders of magnitude.

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