Investigation of the relaxation rate of phonons in the antiferromagnet $FeBO_3$

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Excitation of transverse phonons was achieved in $FeBO_3$ single-crystals by parallel pumping with a vhf magnetic field $h \cos\omega_p t$. The relaxation rate of the phonons excited was measured over a wide range of frequencies, temperatures, and magnetic fields. The above-threshold susceptibility of the system of parametric phonons χ'' was studied, from which it was established that the stationary state of this system is determined by the positive nonlinear relaxation. The dependence of phonon relaxation η on the wave amplitude was obtained: $\eta = \eta_0 + \beta M$, where η_0 is the linear relaxation, M is the total magnetic moment of the parametric phonons, and β is the nonlinear attenuation coefficient. It is shown that β for the phonons agrees as to order of magnitude with β for spin waves. A maximum is observed at $T_c \approx 5$ K in both the relaxation rate η_0 and in the parameter β. Comparison is made between the results and the theory of slow relaxation of phonons by a paramagnetic impurity. The impurity signal was observed by the EPR method, the behavior of which in the region of $T \sim T_c$ indicates a gradual transition of the impurity into a magnetically ordered state.

One of the basic problems in the experimental physics of dielectrics is the study of the spectra and relaxation rates of elementary elastic and magnetic excitations—phonons and magnons. Phonons are usually excited and recorded by transducers stuck to the specimen being studied, the most powerful means of studying the relaxation rate of electron and nuclear magnons being their excitation by the method of parallel pumping by a vhf magnetic field (see reviews, Refs. 1–3). Increasing the vhf magnetic field $h \cos \omega_p t$ above a threshold value h_c induces a parametric instability with respect to the decay of a pumping quantum into two magnons with equal and oppositely directed wave vectors. The critical h_c is proportional to the relaxation rate η of the excited quasiparticles: $h_c = \eta/V$, where V is their coupling coefficient with the pumping field. A wide range of studies of electronic and nuclear magnons has been carried out by the method of parallel pumping.[1-3] Transverse phonons were also excited in the antiferromagnet $FeBO_3$ by this same method.[4] Since the parallel pumping method is contactless, it makes it possible to measure phonon relaxation in a specimen not loaded acoustically on a transducer. Almost ideal conditions are then realized for studying the intrinsic relaxation of phonons in the material.

The aim of the present work was the measurement of the relaxation rate of transverse phonons in an antiferromagnet over a wide range of frequencies, temperatures, and magnetic fields. The linear relaxation of phonons can be determined by measuring the parallel pumping threshold, while a study of the above-threshold magnetic susceptibility of the phonon system makes it possible to elucidate the dependence of the phonon relaxation rate on their number, i.e., to determine the nonlinear relaxation of a system of parametric phonons.

MAGNETOELASTIC INTERACTION IN AN ANTIFERROMAGNET AND PARALLEL PUMPING OF PHONONS

We used the high-temperature antiferromagnet $FeBO_3$ (Néel temperature $T_N = 348$ K) as the object for study, having an "easy plane" type of anisotropy and a weak ferromagnetic moment lying in the plane of easy direction of magnetization. The magnetoelastic interaction leads to a mixing of the phonon and magnon oscillations. As a result the new normal modes of the coupled magnetoelastic oscillations are of the form (see, for example, Ref. 5):

$$\omega_e^2=\omega_{e0}^2+\gamma^2H_\Delta^2,\qquad \omega_{ph}=c(1-\gamma^2H_\Delta^2/\omega_e^2)^{1/2}k, \tag{1}$$

where ω_e and $\omega_{\rm ph}$ are the frequencies of the quasimagnon and quasiphonon branches, $\omega_{e0} = [\gamma^2 H(H + H_D) + s^2k^2]^{1/2}$ is the magnon frequency without taking into account the magnetoelastic interactions, γ is the gyromagnetic ratio, s is the spin-wave velocity, c is the velocity of sound without taking account of the magnetoelastic coupling, k is the wave vector, γH_Δ is the magnetoelastic gap in the magnon spectrum, H_D is the Dzyaloshinskiĭ-Moriya field and H is the static magnetic field in the easy plane. For $k = 10^3$–10^4 cm^{-1} and $H \geqslant 100$ Oe the condition $\gamma^2 H(H + H_D) \gg s^2k^2$ is satisfied and the phonon spectrum can be taken as linear:

$$\omega=c\left[\frac{H(H+H_D)}{H(H+H_D)+H_\Delta^2}\right]k=\tilde{c}k, \tag{2}$$

where the renormalized velocity of sound $\tilde{c}$ depends on the magnitude of the magnetic field.

Thanks to the magnetoelastic interaction there exists a coupling between the alternating magnetic field $h \cos \omega_p t$ and the elastic vibrations of the specimen, and thus a possibility of pumping vhf energy from the magnetic field h into the phonon branch of the oscillations. The connection between the threshold amplitude h_c of the alternating parallel pumping field and the relaxation rate of the excited phonons has been calculated.[6] We obtain from this for the condition $\omega_p \ll \omega_e$

$$\eta_k=Vh_c,\qquad V=\frac{\gamma^2H_DH_\Delta^2\omega_{ph}}{4\omega_e^4}. \tag{3}$$

We previously[4] carried out a detailed experimental test of Eq. (3) at a temperature $T = 77$ K, studying the effect on the threshold amplitude h_c of modulating the phonon energy spectrum. A radiofrequency magnetic field $H_m \cos \omega_m t$ $\times (2\gamma_k \ll \omega_m \ll \omega_p)$ was used for the modulation. As a result,

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the coefficient V was determined experimentally and it was shown that over a wide range of variation in magnetic field and frequency it was described well by Eq. (3). In this way the results of Ref. 4 verify the legitimacy of the use of Eq. (3) for calculation of the relaxation of parametric phonons. The method for measuring h_c has been described before.[4] The specimen is placed in a helical resonator with quality factor $Q_n = 200$–1200. The relative accuracy in measuring h_c was 5% at a fixed frequency and 20% on changing the frequency; the accuracy in the absolute measurement of h_c was 30%. The single-crystal specimens of $FeBO_3$ were naturally faceted plates of thickness 1.2 and 1.3 mm. The growth plane of a plate coincided with the basal crystal plane. Measurements were made over the ranges of temperature $T = 1.5$–293 K, magnetic field $H = 100$–1300 Oe and frequencies $\nu_k = \nu_{p/2} = 250$–770 MHz. The static magnetic field and the vhf pumping field were parallel to one another and to the easy plane of magnetization.

LINEAR RELAXATION OF PHONONS

Measurements of the threshold parallel pumping field were carried out in two specimens. All the functional $h_c(H,T,\nu_p)$ dependences were the same in these specimens, although the actual value of h_c in specimen No. 1 was about one and a half times larger then in specimen No. 2. The frequency and temperature dependences of the threshold field for parametrically excited phonons are shown in Figs. 1–3.

The field dependences of h_c over the whole temperature and pumping frequency range are monotonic functions of H, which can be represented approximately by the law $h_c \propto H^2$. The rather great scatter of the points in Fig. 1 is due to the existence of a size effect in the specimens studied.[4] The problem is that when a whole number of half wavelengths fit into the plate thickness of the specimen, then the Q-factor of such an elastic oscillation is higher and the parallel pumping threshold is correspondingly lower. This effect is particularly strong at nitrogen temperature, when the phonon mean free path is appreciably greater than the specimen thickness.

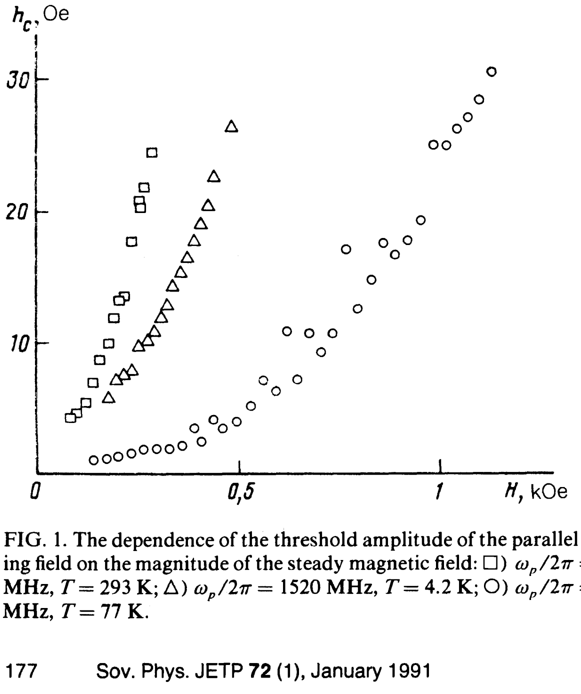

FIG. 1. The dependence of the threshold amplitude of the parallel pumping field on the magnitude of the steady magnetic field: □) $\omega_p/2\pi = 1303$ MHz, $T = 293$ K; △) $\omega_p/2\pi = 1520$ MHz, $T = 4.2$ K; ○) $\omega_p/2\pi = 1290$ MHz, $T = 77$ K.

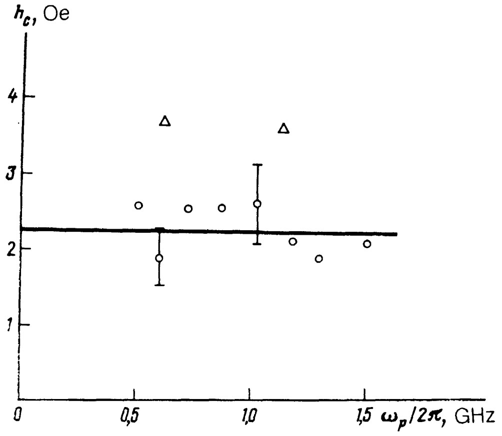

FIG. 2. Frequency dependence of the parallel pumping threshold for two specimens: △) specimen No. 1, ○) specimen No. 2; $T = 77$ K, $H = 330$ Oe.

The investigation of the frequency dependence of the pumping threshold showed (Fig. 2) that within the experimental uncertainty the threshold field remains constant over the whole range of frequency. We note that in experiments on perpendicular pumping of phonons in $FeBO_3$ at a frequency $\nu_p = 35$ GHz the threshold field at $T = 77$ K and $H = 330$ Oe was $h_{c\perp} \approx 2$ Oe (Ref. 7) i.e., a value agreeing with our results. However, parallel pumping was not observed there.[7] This, evidently, indicates that the parallel pumping threshold at a frequency of 35 GHz is appreciably higher than $h_{c\perp}$.

The temperature dependences of h_c at all pumping frequencies and in all magnetic fields have a peak at a temperature $T = 5.3 \pm 0.7$ K (Fig. 3). The threshold peak to the left grows approximately linearly with increase in temperature: $h_c \propto T$. The minimum value of the critical pumping field is reached at a temperature $T \approx 50$–60 K. It is interesting to note that for perpendicular pumping of phonons and parallel pumping of magnons[7] in $FeBO_3$ at a frequency $\nu_p = 35$ GHz, the temperature dependences of the threshold had a peak at $T = 14 \pm 4$ K. Since the nature of this peak is evidently one and the same in all experiments, it can be assumed

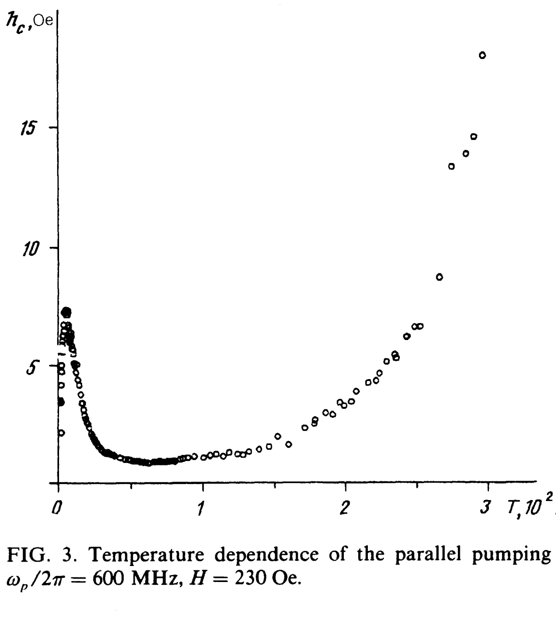

FIG. 3. Temperature dependence of the parallel pumping threshold; $\omega_p/2\pi = 600$ MHz, $H = 230$ Oe.

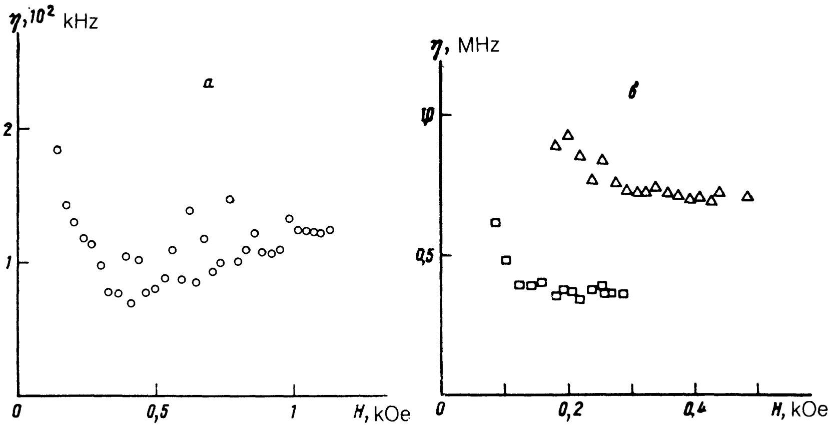

FIG. 4. Dependence of the phonon relaxation rate on the magnitude of the steady magnetic field: (a) O) $\omega_p/2\pi = 1290$ MHz, $T = 77$ K; (b) □) $\omega_p/2\pi = 1303$ MHz, $T = 293$ K and Δ) $\omega_p/2\pi = 1520$ MHz, $T = 4.2$ K. Equation (3) was used in the calculation with the following values of the parameters: $H_\Delta^2 = 3.9$ kOe^2 and $H_D = 101$ kOe for $T = 4.2$ K; $H_\Delta^2 = 3.4$ kOe^2 and $H_D = 99$ kOe for $T = 77$ K; $H_\Delta^2 = 0.5$ kOe^2 and $H_D = 64$ kOe for $T = 293$ K.

that the temperature of the peak depends weakly on frequency: on raising the pumping frequency about 30-fold the temperature of the peak rises about 3-fold.

When evaluating the phonon relaxation rate by Eq. (3) we used values of the Dzyaloshinskiĭ-Moriya field H_D measured by Velikov *et al.*[8] and values of the magnetoelastic constants H_Δ^2 measured by us from observation of the size effect (see Ref. 4 for the method of determining H_Δ^2 from the size effect). By observing the size effect for different specimen temperatures we determined both the magnetic field and temperature dependence of the velocity of sound and calculated the value of H_Δ^2. Comparison of the values of H_Δ^2 obtained with the results of Velikov *et al.*[8] shows that the value of H_Δ^2 in our specimens is about 20% less, while the temperature dependences are identical.

Values of the phonon relaxation rate as a function of the magnitude of the magnetic field are shown in Fig. 4 for three temperatures. The values of H_D and H_Δ used for the calculation are shown in the figure caption. In fields $H \approx 100$ Oe an increase in phonon relaxation sets in with decreasing field, in the remaining magnetic field interval the phonon relaxation depends weakly on the magnetic field. A similar result was obtained by Kotyuzhanskiĭ and Prozorova[7] for perpendicular pumping of phonons: on increasing H from 250 to 1200 Oe the relaxation increased about 1.25-fold.

The dependence of the relaxation rate on the pumping frequency is shown in Fig. 5 for two specimens of $FeBO_3$. It turned out that the relaxation rate is proportional to the pumping frequency over the whole frequency range. Since for fixed values of the static magnetic field and temperature, and for $k < 10^5$ cm^{-1}, the relation $\omega_{ph} = \text{const}\cdot k$ holds to high accuracy, the plots in Fig. 5 can also be interpreted as proportionality of the relaxation rate to the wave vector of the excited phonons: $\eta \propto k$.

The temperature dependence of the phonon relaxation rate is shown in Fig. 6 for fixed values of H and ω_p. We note that such a functional dependence $\eta(T)$ is observed for all values of H and ω_p. The relaxation rate at the peak for $T = 5.3 \pm 0.7$ K is about 10 times greater than minη corresponding to a temperature $T \approx 60$ K. For $T < 5$ K the relaxation rate is almost proportional to the temperature; for 250 K $> T >$ 60 K a nearly linear growth in relaxation is observed, which is described by the empirical law $\eta_1 = 18$ kHz $+ T$ [K] $\times 0.205$ kHz/K.

Increasing the temperature further ($T > 250$ K starts a rapid growth in the relaxation parameter calculated accord-

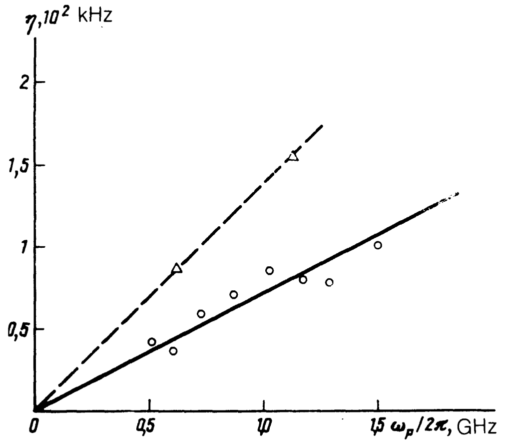

FIG. 5. Dependence of phonon relaxation rate on the pumping frequency for two specimens: Δ) specimen No. 1, O) specimen No. 2; $T = 77$ K, $H = 330$ Oe.

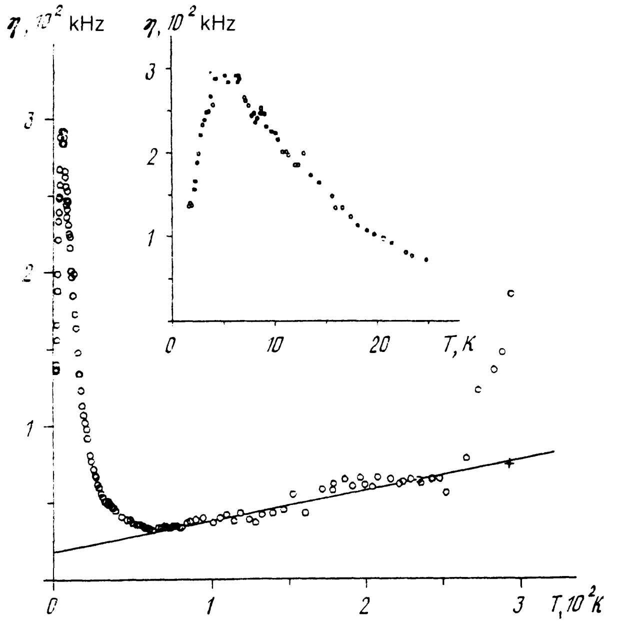

FIG. 6. Temperature dependence of phonon relaxation rate in specimen No. 2 for $\omega_p/2\pi = 600$ MHz, $H = 230$ Oe; O) calculated according to Eq. (3), +) measured by the modulation method. The straight line is a plot of the empirical formula for η_2 (see text).

ing to Eq. (3). However, in the course of further experiments it was made clear that the phonon relaxation rate in this temperature range is evidently described by the same linear law for η_1 as for $T<250$ K, and the theoretical equation (3) simply ceases to be satisfied as the temperature approaches $T_N = 348$ K. Our doubts about the applicability of Eq. (3) at high temperatures first arose as a result of the size effect at $T=293$ K being observed about as well as at $T=77$ K, although according to the calculation $\eta = h_c V$, the mean free path at $T=293$ K was appreciably reduced and became about equal to the specimen thicknesses, which should lead to a weakening of the influence of the boundaries on the pumping threshold. We therefore carried out measurements of η at $T=293$ K by another, independent, method, namely by the influence of the modulating field $H_m \cos \omega_m t$ on the threshold.

The effect of the modulation of the phonon spectrum on the threshold for their excitation h_c, for the case $\omega > 2\eta$ is given by the formula[9]

$$\frac{h_c}{h_{c0}}-1=\frac{4U^2H_m^2}{\omega_m^2+(2\eta)^2}, \tag{4}$$

where U is the coupling coefficient of the modulating field with the phonon system ($U=V$) and h_{c0} is the pumping threshold for $H_m=0$. We obtain from Eq. (4)

$$\left[\frac{h_c}{h_{c0}}-1\right]^{-1}\propto\omega_m^2+(2\eta)^2. \tag{5}$$

Measurements of the dependence of h_c/h_{c0} on ω_m at $T=293$ K yielded the value $\eta = 75 \pm 25$ kHz (this value is shown by a cross in Fig. 6), which is less by more than a factor of two than the value of η calculated from Eq. (3). Substituting $\eta = 75$ kHz into Eq. (4) gives $U_{\text{exp}} = (5.2 \pm 1)\cdot 10^4$ Hz/Oe.

Using the experimental values $\eta = 75$ kHz and $h_c = 12$ Oe at $T=293$ K and $H=190$ Oe, we obtain $V_{\text{exp}} = (1.6 \pm 0.8)\cdot 10^4$ Hz/Oe. Theory, however, gives $V=U=(3.6\pm 0.7)\cdot 10^4$ Hz/Oe. It can thus be stated that the use of Eqs. (3) and (4) at $T=293$ K leads to the relation $V_{\text{exp}} \neq U_{\text{exp}}$, with the coefficient U_{exp}, i.e., the coupling between the phonons and the low-frequency field H_m agreeing with the calculated value, to within the accuracy, while $V_{\text{exp}} = \eta/h_0$ is appreciably less than the calculated V_{th}.

These results can be explained by the increase in fluctuations of the magnetic moment of the specimen on approaching the temperature of the antiferromagnetic transition point T_N. Fluctuations modulate the spin-wave and phonon spectra by means of noise. This leads to a raise of the pumping threshold and to the destruction of the relation $h_c V = \eta$. We note that LeCraw *et al.*[10] observed for $T>250$ K a nonlinear increase in the antiferromagnetic resonance line width in $FeBO_3$. These results agree qualitatively with ours.

We now return to the discussion of phonon relaxation. On perpendicular pumping at a frequency $\nu_p = 35.4$ GHz (in the range 1.2 K $<T<180$ K) phonon relaxation had a temperature dependence similar to Fig. 6. However, the peak was observed at $T\approx 14$ K[1)] and the relaxation peak rose about 2–3-fold and not 10-fold as in our experiments. We note that an estimate[7] of the absolute value of the phonon relaxation rate gave a value $\eta \approx 1$ MHz at $T=90$ K, $H\to 0$, i.e., an order of magnitude greater than in our experiments.

According to theoretical calculations the temperature peak in the phonon and magnon relaxation can be explained by a process of slow relaxation at a paramagnetic impurity.[11] If there are in the crystal local impurity levels with activation energies $E \gg \hbar\omega_{\text{ph}}$ and relaxation time τ_i, spin waves and quasiphonons lead to modulation of the impurity levels and of their population. The population change is delayed relative to the change in position of the levels due to the finite relaxation rate of an impurity: $\gamma_\parallel = \tau_i^{-1}$. The attenuation of magnetic oscillations by the slow relaxation process is a maximum for the case $\omega_{\text{ph}} = \gamma_\parallel$. If we assume that the relaxation time τ_i is described by the Arrhenius formula

$$\tau_i=\tau_0\exp(E/k_BT), \tag{6}$$

we obtain the following dependence of the temperature T_c of the peak on the phonon frequency:

$$T_c^{-1}=\frac{\ln\omega}{E}k_B-\frac{\ln\tau_0}{E}k_B. \tag{7}$$

This relation should be described by a straight line with slope k_B/E on a $\ln \omega - T_c^{-1}$ plot, with the intersection of the line with the ordinate axis giving the value of $(k_B \ln \tau_0)/E$. In this way, having the $T_c(\omega)$ dependence, both the activation energy of the impurity E and the time for its relaxation τ_0 can be determined. Since the logarithmic dependence is very weak, the values of T_c obtained in the phonon frequency range we studied do not allow us to construct this dependence—the error in measuring T_c is greater than its change. However, by using together with our results those of Kotyuzhanskiĭ and Prozorova[7] at a frequency $\nu_{\text{ph}} = 2\pi\cdot 17.7$ GHz and of Kotyuzhanskiĭ *et al.*[12] who studied the Q-factor of different modes of intrinsic magnetoelastic oscillations, we obtain the $T_c^{-1}(\ln\omega)$ dependence shown in Fig. 7. The best fit to the experimental points is given by a straight line with the parameters $E/k_B = 36$ K, $\tau_0 = 0.8\cdot 10^{-12}$ s.

This estimate of E and τ is highly approximate. Mikhaĭlov and Farzetdinova[13] calculated the slow relaxation much more rigorously and obtained the following expression for η:

$$\eta=-2c\left[\frac{\Phi}{\hbar}\right]^2\gamma_\parallel(E)I'\frac{\omega_k}{\gamma_\parallel^2(E)+\omega_k^2}, \tag{8}$$

where c is the impurity concentration, $I=[\exp(E/k_BT)+1]^{-1}$ is the thermal population of an impurity level, $I'=dI/dE$, Φ is the amplitude of the interaction between a parametric phonon and a two-level impurity, and $\gamma_\parallel$ is the relaxation rate of a two-level impurity. The interaction amplitude Φ for $T \ll T_N$ is almost independent of temperature, meaning that the position in temperature of the peak T_c will be determined by the temperature dependence $I'(T)$ and $\gamma_\parallel$. Under this condition the maximum for η will not be of the form $\omega = \gamma_\parallel$, but the magnitude of T_c changes as the function $I'(T)$ allows. Curve 1 in Fig. 7 shows the frequency dependence of T_c, obtained from Eq. (8) under the condition that $\gamma_\parallel$ is given by the Arrhenius law (6) with the parameters $E/k_B = 35$ K and $\tau_0 = 0.25\cdot 10^{-12}$ s.

The contribution to the relaxation of a two-level impurity from single-particle emission processes by an impurity of a resonance magnon or phonon with energy $\hbar\omega = E$ was considered by Mikhaĭlov and Forzetdinova.[14] According to their calculations,[14] for such a process

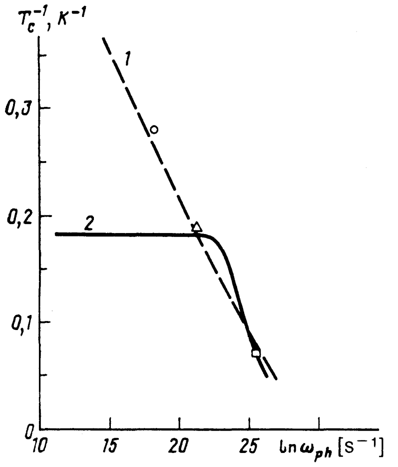

FIG. 7. Frequency dependence of the temperature of the peak in phonon relaxation in (T_c^{-1}, $\ln \omega_{ph}$) coordinates.The experimental values are taken from the following work: O) Kotyuzhanskiĭ and Prozorova,[12] △) present work, □) Kotyuzhanskiĭ and Prozorova.[7] Dashed curve 1 is calculated from Eqs. (6) and (8). Full curve 2 corresponds to the value of T_c obtained from Eqs. (8) and (9).

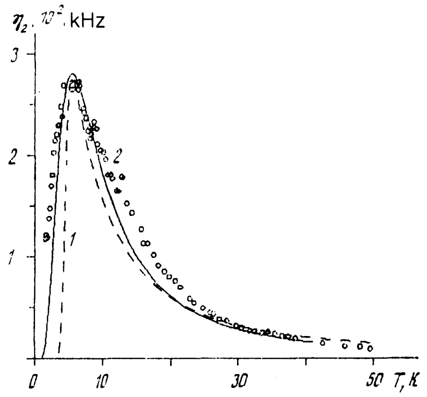

FIG. 8. Temperature dependence of the phonon relaxation parameter $\eta_2 = \eta - \eta_1$ for $\omega_p/2\pi = 600$ MHz, $H = 230$ Oe. The dashed curve 1 is calculated from Eq. (8) with the impurity relaxation in the form of Eq. (6). The full curve 2 is calculated according to Eq. (8) with the impurity relaxation in the form of Eq. (9).

$$\gamma_{\parallel} = \tau_0^{-1} \operatorname{cth}(E/2k_BT). \tag{9}$$

Substituting this function into Eq. (8) we find the $T_c^{-1}(\omega_p)$ relation for the case when the contribution of an impurity to the relaxation comes from single-particle transition processes with the emission of a magnon or phonon. Curve 2 of Fig. 7 corresponds to this dependence, shown with the parameters $E/k_B = 11$ K, $\tau_0 = 0.65\cdot 10^{-10}$ s. This choice of parameters makes it possible to describe the points for $T \geqslant 5.3$ K, but does not allow the description of the further lowering of T_c on reducing the frequency to $\nu_{ph} = 13$ MHz, since T_c reaches a constant. If we take $E/k_B = 7$ K, so that curve 2 should become constant at $T \approx 3.5$ K, then for $\nu_{ph} \approx 300$ MHz its peak will still occur at $T_c \approx 3.5$ K, while the $\eta(T)$ temperature dependence for $\nu_{ph} = 17.7$ GHz has practically no peak, since for $T_c > E/k_B$ the peak flattens out very quickly.

Finally, if the sum of the Arrhenius contributions and single-particle processes are taken into account in $\gamma_{\parallel}$ of an impurity, we obtain a $T_c^{-1}(\omega_{ph})$ dependence more like curve 2, i.e., the curve nevertheless reaches $T_c = \text{const}$ and does not describe all the experimental points.

So far we have mainly discussed the position of the $\eta(T)$ peak, i.e., the function $T_c(\omega_{ph})$, but the dependence of slow phonon relaxation on temperature, magnetic field and phonon frequency is no less interesting. Values of the relaxation $\eta_2 = \eta -$ (18 kHz + T[K]$\cdot$0.205 kHz/K) are shown in Fig. 8, obtained by subtracting from the experimental results for the total phonon relaxation the linear contribution η_1. The theoretical $\eta(T)$ dependence for slow phonon relaxation is also shown in Fig. 8 when the main contribution to impurity relaxation is made by the Arrhenius formula (Eq. 6) (curve 1) and by Eq. (9) for single-particle emission by magnon or phonon admixture (curve 2). At temperatures $T > T_c$ both curves describe the experiment equally well. For $T < T_c$ the use of the Arrhenius formula and Eq. (8) gives an exponential growth in relaxation $\eta(T)$ and does not fit the experimental results. If we use Eq. (9) for $\gamma_{\parallel}$, Eq. (8) (see curve 2) describes the results in the region $T < T_c$ fairly well.

We will now dwell on the frequency and field dependences of phonon relaxation. Experiment shows that $\eta \propto \nu_{ph}$ and depends weakly on H, which means that the amplitude Φ of the interaction of a phonon with a two-level impurity in Eq. (8) should be practically independent of ν_{ph} and H. The interaction of phonons with an impurity is due both to pure elastic anharmonicity and to magnetoelastic, relativistic, and exchange interactions. In order to describe the experimental $\eta(\nu_{ph}, H)$ dependences by a slow phonon relaxation process one must choose in the theoretical analysis of the amplitude Φ that interaction (combination of interactions) which, over the range of the experimental parameters, makes Φ independent of the phonon frequency and of the magnetic field.

Lutovinov[15] made an attempt to attribute the existence of a peak in the $\eta(T)$ dependence not to a slow relaxation process but to a three-particle process of coalescence of a phonon with a magnon into a magnon. The presence of an impurity in the specimen can in principle lead to an increase in the amplitude of such an interaction, especially in the region of $k_BT \approx E/2$, which would lead to the appearance of a peak on the $\eta(T)$ dependence. Lutovinov[15] obtained the following expression for phonon relaxation in such a three-particle process:

$$\eta \propto \frac{\nu_{ph} T}{H^{5/2}} \operatorname{th} \frac{\nu_{ph} T}{H^{5/2}}. \tag{10}$$

This formula describes the $\eta(T)$ dependence quite well for fixed pumping frequency and $E/k_B = 11$ K, and also the growth in $\eta \propto \nu_{ph}$. However, it does not explain the shift in T_c with chance frequency and mainly predicts a very strong dependence of phonon relaxation rate on magnetic field $\eta \propto H^{-5/2}$, which is not observed in the experiment.

ELECTRON PARAMAGNETIC RESONANCE OF IMPURITY IONS IN $FeBO_3$

Since all the theoretical models which we have used to explain the low-temperature relaxation peak are based on a consideration of the interaction of magnons and phonons with an impurity, we carried out additional investigations aimed at finding impurities in $FeBO_3$ specimens. Using an ESP-300 spectrometer made by the Bruker firm, we studied

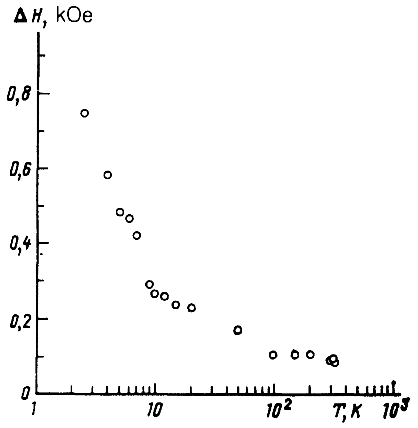

FIG. 9. Temperature dependence of the EPR linewidth of the paramagnetic impurity for $\omega_p/2\pi = 9.45$ GHz.

EPR signals of frequency $\nu_p = 9.45$ GHz at temperatures 2.5–440 K. At $T > T_N = 348$ K strong EPR signals were observed from paramagnetic Fe^{3+} with a Landé factor $g = 2.00$. For $T < T_N$, beside the antiferromagnetic resonance signal, a weak EPR signal likewise with $g = 2.00$ was observed, which could be interpreted as a signal from a paramagnetic impurity. The ratio of the attenuation of the vhf signal by the impurity at $T = 310$ K and by Fe^{3+} ions at $T = 360$ K equals $I(T = 310\ \mathrm{K})/I(T = 360\ \mathrm{K}) = 1.6 \cdot 10^{-4}$. If it is assumed that the susceptibility of the impurity ion is approximately equal to the susceptibility of Fe^{3+} ions, then the ratio obtained characterizes the impurity concentration. However, this assumption has not been verified so far, since it is not known on exactly which impurity ions the paramagnetic resonance signal is observed.

On lowering the temperature an increase in the impurity EPR line-width ΔH takes place (Fig. 9) and the line width increases by an order of magnitude. At $T \approx 10$ K, beside the broadening, a shift in the impurity EPR line to smaller field also starts (Fig. 10).

The behavior of the EPR impurity line at low temperatures is thus similar to the EPR of Fe^{3+} at $T > T_N$, i.e., near the antiferromagnetic ordering temperature. However, the transition in the impurity system is strongly spread out, which could be due to a non-uniform distribution of the impurity throughout the specimen.

Since the maximum of the phonon relaxation lies approximately in the same temperature interval in which the behavior of the impurity system differs from paramagnetic, evidently the paramagnetic impurity observed by us is due to slow phonon relaxation in $FeBO_3$. We also note that all the

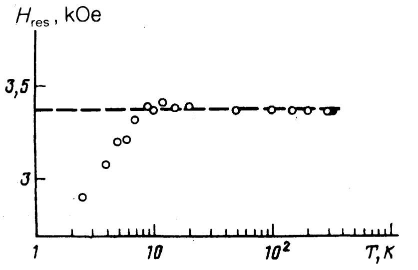

FIG. 10. Temperature dependence of the position of the impurity EPR line at $\omega_p/2\pi = 9.45$ GHz.

results given were obtained in a magnetic field parallel to the basal plane. Rotation of the magnetic field from a plane to the $\mathbf{C}_3$ axis does not lead to a shift of the impurity EPR line, but reduces its intensity practically to zero.

INVESTIGATION OF THE STATIONARY STATE OF A SYSTEM OF PARAMETRIC PHONONS

Investigations of the stationary state of a system of parametric magnons have been carried out since the start of the 60's, i.e., from the time that Schlömann *et al.*[16] discovered parametric excitation of magnons in a ferromagnet by the method of parallel pumping. The simplest method for investigating the stationary state is the measurement of the nonlinear high-frequency susceptibility $\chi = \chi' + i\chi''$ as a function of the excess vhf magnetic field h above its threshold value h_c. The cessation of the exponential growth of the amplitude of the parametric wave and the assumption of a stationary value by the amplitude is due, according to theoretical ideas, to the following reasons.

1. A nonlinear increase in relaxation beyond the threshold of parametric excitation.[17] In the simplest case we can write $\eta = \eta_0 + \varkappa N$, where η_0 is the linear relaxation ($\eta_0 = h_c V$), N is the number of parametric magnons, $\varkappa$ is the coefficient of the positive nonlinear attenuation. For fixed amplitude of the vhf field h ($h > h_c$) the value of N grows until the relation $\eta_0 + \varkappa N = hV$ becomes satisfied, characterizing a stationary state in the parametric system with nonlinear susceptibility

$$\chi' = \chi_0, \quad \chi'' = \frac{4V^2}{\varkappa}\left[\frac{h - h_c}{h}\right] = \frac{4V^2}{\varkappa}\left[\frac{\xi - 1}{\xi}\right], \tag{11}$$

where $\xi = h/h_c$ is the supercriticality and χ_0 is the linear dynamic susceptibility [for $FeBO_3$ $\chi_0 \approx \chi_\perp = 1.6 \cdot 10^{-4}$ (Ref. 18)].

2. A phase mechanism that limits the number of parametric waves.[1] It is characterized by a phase shift of a parametric pair of waves relative to the phases of the pumping field until the relation $hV \sin \psi_k = \eta_0$ is satisfied. In that case the stationary state is characterized by the susceptibility

$$\chi' = \chi_0 + \frac{2V^2}{S} \cdot \frac{\xi^2 - 1}{\xi^2}, \qquad \chi'' = \frac{2V^2}{|S|} \frac{(\xi^2 - 1)^{1/2}}{\xi^2}; \tag{12}$$

here S is the four-particle interaction constant. Although both methods for limiting the amplitude act in a real experimental situation and it is necessary to use the condition

$$\eta_0 + \varkappa N = hV \sin \psi_k,$$

nevertheless, comparison of the experimental $\chi(\xi)$ dependence with the relations (11) and (12) allows one as a rule to understand which is the main mechanism and estimate $\varkappa$ or S.

After the discovery of parametric resonance of magnons in antiferromagnets by Seavey[19] and by Prozorova and Borovik-Romanov,[20] corresponding investigations of χ' and χ'' were also carried out in ferromagnets, including $FeBO_3$ (Ref. 18). The measurements showed that the main limitation mechanism in $FeBO_3$ of the number of parametric magnons with frequency $\nu_{ph} = 17.7$ GHz at temperatures $T = 1.6$–2.2 K is positive nonlinear relaxation.

The present work is the first study of the nonlinear magnetic susceptibility for parametric phonon resonance. The method of parallel pumping is described above. The measurements of susceptibility were carried out at several frequencies in the range $\nu_p = 650$–1450 MHz at temperatures 1.5–293 K in magnetic fields $H = 150$–700 Oe. The above-threshold susceptibility was measured by determining the change in resonance characteristic of the resonator, which takes place on parametric excitation of phonons in the specimen. The change in natural frequency of the resonator is associated with a change in the magnitude of χ', while the worsening of its Q-factor is connected with the appearance of χ'':

$$\chi'=\chi_0+\frac{1}{2\pi A}\frac{\omega-\omega_0}{\omega_0},$$
$$\chi''=\frac{2\pi}{A}\left[\frac{1}{Q_n}-\frac{1}{Q_{0n}}\right]=\frac{1}{4\pi Q_{0n}}\left[\left[\frac{P_{\rm in}}{P_{\rm out}}\right]-1\right], \quad (13)$$

where

$$A=\int_{V_{\rm spec}} h^2\,dV \Big/ \int_{V_{\rm res}} h^2\,dV$$

is the filling factor, i.e., the ratio of the energy stored in the specimen to the field energy stored in the resonator, $V_{\rm spec}$ and $V_{\rm res}$ are the volumes of specimen and resonator, ω_0 and ω are the natural frequencies of the resonator before and after excitation of the parametric waves, Q_{0n} and Q_n are the Q-factors of the loaded resonator before and after excitation of the parametric waves, $P_{\rm in}$ and $P_{\rm out}$ are powers at the input and output of the resonator measured in units of the threshold power. For measurement of these quantities, precision attenuators were fitted to the input and output of the resonator and maintained a constant signal at the crystal detector, which was the terminal component of a vhf circuit. Readings from both attenuators were read (this method for measuring χ'' has been described in detail by Zautkin[21]). The accuracy of a relative measurement of χ'' at fixed pumping frequency was not worse than 10%.

To prevent heating of the specimen we worked with short pulses of duration $\tau = 100$–$300\,\mu$s and off-duty factor 100–1000. In addition, estimates were made of the maximum heating of the specimen in the course of one pulse under the assumption that adiabatic heating of the specimen takes place during the time of a pumping pulse. Taking for the estimate the heat capacity at nitrogen temperature $C \sim 40$ J/mole·deg, the experimental value of χ'' and of the attenuated power in the form $P_{\rm att} = \omega_p \chi'' h^2 V_{\rm spec}/2$, we obtain the heating of the specimen during the time $\tau = 300\,\mu$s to be 10^{-3} K. To eliminate the possibility of heating the specimen by repeated action of pumping pulses, we carried out control experiments in which one junction of a thermocouple was fixed to the specimen. It was established that noticeable heating of the specimen placed in helium gas at $T = 77$ K and 293 K starts when the off-duty factor becomes less than 10. For measuring χ'' at helium temperatures the resonator was filled with superfluid helium.

Characteristic dependences of χ'' on the supercriticality are shown in Fig. 11 for two values of the magnetic field. The theoretical $\chi''(\xi)$ dependence corresponding to the two mechanisms for limiting the amplitude of parametric waves are shown by curves 1 and 2 in Fig. 11: 1 is calculated according to Eq. (11) for nonlinear relaxation, and 2 according to Eq. (12) for the phase mechanism for the limiting. The absence of theoretical calculations of the coefficient of four-particle interaction S only allows us to make a comparison of the experimental results with the theory of the phase mechanism of limitation according to the functional $\chi''(\xi)$ dependence, since neither the absolute value of the coefficient S nor its functional dependence on T, ω, and H are known. However, having the functional $\chi''(\xi)$ dependence, allows us to conclude that the main contribution to limiting the number of parametric phonons comes from the mechanism of positive non-linear relaxation (curve 1 of Fig. 11).

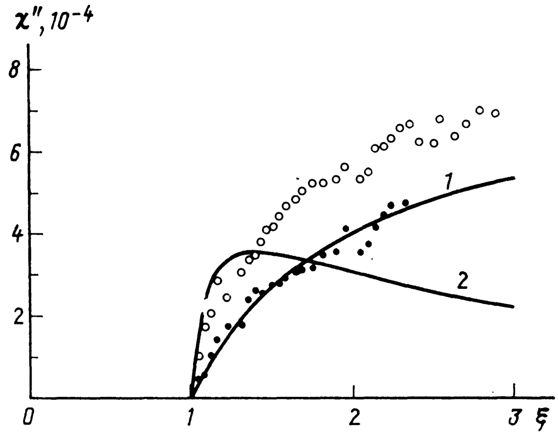

FIG. 11. Nonlinear magnetic susceptibility χ'' of phonons as a function of the excess of the pumping field amplitude above threshold $\xi = h/h_c$: ○) $H = 266$ Oe; ●) 496 Oe; $\omega_p/2\pi = 1450$ MHz, $T = 77$ K. Curve 1—calculated according to Eq. (11). Curve 2—calculated according to Eq. (12).

Beside the functional relations shown in Fig. 11 an investigation of the value of $\Delta\chi'$ can act as confirmation of this conclusion. In our experiments the change in χ' by parametric excitation of phonons, which the theory of positive nonlinear relaxation also predicts, was not observed, so that we can only estimate an upper limit for $\Delta\chi'$, starting from the experimental accuracy and comparing it with that calculated from Eq. (12). It follows from Eq. (12) that for the phase mechanism of limitation $\Delta\chi' = \chi''(\xi^2-1)^{1/2}$, i.e., for the maximum values $\xi \approx 3.5$ reached in the experiment we obtain $\Delta\chi' \approx 3.3\chi''$. An estimate of the upper limit of $\Delta\chi'$ from experiment gives under the same conditions $\Delta\chi' < 1.5\chi''$. This result is yet another argument in favor of the conclusion that the phase mechanism is not the main one in the limiting of the number of parametric phonons.

The dependence of the magnitude of χ'' on the static magnetic field for a fixed excess above threshold is shown in Fig. 12. We observe a dependence close to $\chi'' \propto H^{-1}$. We note that χ'' for a system of parametric magnons[18] grows with increasing static magnetic field. The next figure (Fig. 13) shows the dependence of the coefficient of nonlinear attenuation $\varkappa$ on the magnetic field, calculated from the results in Fig. 11 according to Eq. (11). This dependence is considerably stronger than that of $\chi''(H)$ and is represented approximately by the function $\varkappa \propto H^{-5/2}$. We carried out measurements of χ'' for values of the temperature 1.5, 77 and 293 K and different pumping frequencies. In all these measurements the functional $\chi''(H)$ dependence was the same, i.e., the chief mechanism for limiting the number of parametric phonons was always positive nonlinear relaxation. Calculation of the coefficient $\varkappa$ shows that on reducing the tem-

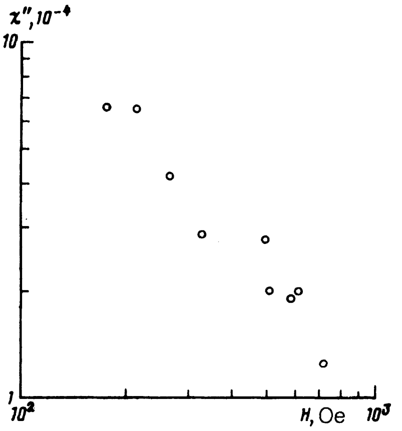

FIG. 12. Dependence of the nonlinear magnetic susceptibility χ'' of phonons on the steady magnetic field; $T = 77$ K, $\omega_p/2\pi = 1450$ MHz, $\xi = 1.5$.

perature from 77 to 1.5 K the nonlinear relaxation increases by as many times as the linear. However, at $T = 293$ K, when η_0 is also noticeably greater than for $T = 77$ K, the value of $\varkappa$ is 4–5 times smaller than at $T = 77$ K. With increase of pumping frequency the parameter $\varkappa$ increases approximately like $\varkappa \propto \omega_{ph}^2$.

Since the above results are for the first experimental study of the nonlinear magnetic susceptibility of parametric phonons and this phenomenon has so far not been studied theoretically, they can be compared only with results of studies of χ'' of a system of parametric spin waves in $FeBO_3$ at a frequency $\nu_p = 35.4$ GHz.[18] If the results of Kotyuzhanskiĭ *et al.*[12,18] on χ'' and on the relaxation of magnons are recalculated to give the quantity $\varkappa$, we get for $T = 1.55$ K, $H = 200$ Oe, $\nu_p = 35.4$ GHz and $\eta_m = 1.1$ MHz the following estimate: $\varkappa_m \approx 5 \cdot 10^{-10}$ Hz·cm$^3 \approx 10^3 \varkappa_{ph}$. It would appear that there is nothing in common between the coefficients of nonlinear relaxation of magnons and phonons—they differ by three orders of magnitude. In actual fact there is something in common and this will be shown in what follows. We will first make some estimates concerning parametric phonons. Using the values of χ'' and $\varkappa$ we find that for a sufficient excess above threshold $\xi \approx 3$, when the nonlinear addition to the relaxation is double the linear contribution,

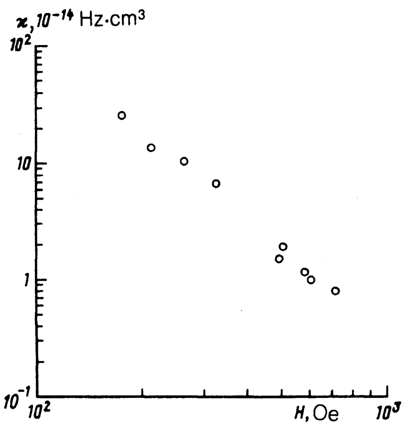

FIG. 13. The coefficient of nonlinear attenuation of phonons as a function of the magnitude of the steady magnetic field; $T = 77$ K, $\omega_p/2\pi = 1450$ MHz, $\xi = 1.5$.

$2\eta_0 = \varkappa N$, the density of parametric phonons is $N \approx 2 \cdot 10^{18}$ cm^{-3}. The energy of a parametric wave corresponding to this value of N is $E_{tot}/V = 10$ erg/cm^3.

From the value of the total energy we can estimate the amplitude of the deformation and the amplitude of the oscillation of the magnetization in the parametric wave. For this purpose we write the total energy in the form[5]

$$E = E_M + E_{MEl} + E_{El},$$

i.e., as a sum of magnetic, magnetoelastic and elastic energies. Keeping the principal terms in the expressions for these energies, we obtain

$$E = MH_D + Bul + cu^2. \tag{14}$$

Here M is the magnetic moment of the system of parametric phonons, u is the deformation of the crystal in the wave, $B = 10^7$ erg/cm^3 is the magnetoelastic and $c \approx 10^{12}$ dyne/cm^2 is the elastic constant of the material. Comparison of the second and third terms shows that $E_{El} \propto u^2$ is comparable to $E_{MEl} \propto u$ for a deformation $u \approx 10^{-5}$, i.e., for $E_{MEl} = E_{El} \approx 100$ erg/cm^3. Since in our experiments the maximum total energy density is an order of magnitude less and $E_{MEl} < E_{tot}$, the term E_{El} can always be neglected.

The first two terms in Eq. (14) are linear in $N (M \propto N$ and $u \propto N)$ so that their relative contribution to the total energy is independent of N and depends only on the magnetic moment of a phonon. We shall find the condition for which $E_M = E_{MEl} = E_{tot}/2$. We rewrite it in the form

$$N\mu_{ph}H_D = N\hbar\omega_{ph}/2,$$

where μ_{ph} is the magnetic moment of a phonon. Substituting in this

$$\mu_{ph} = \hbar \frac{d\omega_{ph}}{dH} = g\mu_B \frac{H_D H_\Delta^2 \omega_{ph}}{4\gamma (HH_D + H_\Delta^2)^2}, \tag{15}$$

where μ_B is the Bohr magneton, we find that for $T = 77$ K the relation $E_M = E_{MEl}$ is satisfied for $H \approx 1266$ Oe. If $H < 1266$ Oe, then $E_M > E_{MEl}$. In particular, in a field $H = 266$ Oe we obtain $E_{MEl} \sim E_M/20$, i.e., $E_{MEl} \sim 0.05 E_{tot}$, $E_M \approx 0.95 E_{tot}$. Taking $B \approx 10^7$ erg/cm^3 as the magnetoelastic constant as an estimate, we obtain $u_{max} \approx 10^{-7}$. This is a fairly weak deformation at which the intrinsic nonlinearity of the elastic system of a crystal generally does not appear. At the same time, as was shown above, because of the strong magnetoelastic coupling these elastic oscillations are accompanied by intense oscillations of the specimen magnetization, $M_{max} \approx 2 \cdot 10^{-3}$ G. In order to provide a static magnetic moment equal to M_{max} a field $H \approx 2$ G is required.

Usually the nonlinearity of a magnetoelastic system is much larger than of an elastic one and it can be suggested that the reason for nonlinear relaxation of phonons is an increase in the amplitude of oscillations in the magnetic moment of the specimen. In that case the formula for the nonlinear attenuation of phonons should be rewritten in the form $\eta = \eta_0 + \beta M$, where $M = N\mu_{ph}$ and $\beta = \varkappa/\mu_{ph}$ is the coefficient of nonlinear relaxation of oscillations of the magnetic moment of the phonons. In what follows we will discuss the magnitude of β and its fundamental dependences. According to Eq. (15) the phonon magnetic moment for $HH_D \gg H_\Delta^2$ is $\mu_{ph} \propto H^{-2}$, consequently conversion of the

TABLE I. Values of the coefficient of positive nonlinear attenuation of magnetization β (in 10^{-12} cm^3·G) for quasiphonons (results of the present work) and quasimagnons (according to Kotyuzhanskiĭ and Prozorova[18]).

$\omega_p/2\pi$, MHz	T, K 1,55	77	293
	Quasiphonons		
1450	6	2,1	1,8
766	–	1,3	–
650	–	1,2	–
	Quasimagnons		
35,4	23 (H⊥C$_2$) 70 (H‖C$_2$)	–	–

$\varkappa(H)$ dependence into $\beta(H)$ will give $\beta \propto H^{-1/2}$, i.e., a fairly weak field dependence of β. The temperature and frequency dependences of the nonlinear relaxation also change. In addition, our results now will be quite comparable with the results of Kotyuzhanskiĭ and Prozorova.[18]

The point is that for $FeBO_3$ usually $\mu_{ph} = (0.01–0.1)\mu_B$, while the magnetic moment of a spin wave $\mu_m \approx 10\ \mu_B$. Thus, conversion for the nonlinear relaxation $\varkappa$ of phonons and magnons to the coefficient β shows that for magnetization oscillations of the quasiphonon and quasimagnon branches of the spectrum the nonlinear attenuation coefficient of the magnetic moment β is of the same order of magnitude (see Table I). Two values of β are shown in Table I for magnons, corresponding to different directions of the magnetic field in the basal plane of the crystal. The growth of β of the phonon branch can be seen in the table in the region of the temperature of the peak in the phonon relaxation and the growth of β on increasing the pumping frequency.

CONCLUSIONS

1. The linear relaxation η_0 of transverse phonons in the antiferromagnet $FeBO_3$ was measured in a wide range of frequencies, temperatures, and magnetic fields. It has been shown that the theory of slow relaxation of phonons by a paramagnetic impurity describes satisfactorily the experimental results in the low temperature region.

2. An EPR signal which can lead to slow relaxation of phonons and magnons was observed in a $FeBO_3$ single crystal.

3. The nonlinear magnetic susceptibility of parametric phonons was measured for the first time. It is shown that the stationary state of parametric phonons in the antiferromagnet $FeBO_3$ is determined by the mechanism of positive nonlinear attenuation.

4. It is established that the coefficients of positive nonlinear relaxation of oscillations of magnetization of the quasiphonon and quasimagnon branches of the spectrum in $FeBO_3$ are of the same order of magnitude.

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[1)] In the text of Ref. 7 the temperature of the peak is given as $T_c = 18$ K, but in Fig. 4 there the relaxation peak corresponds to $T_c = 14 \pm 4$ K.

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