Parallel pumping of phonons in antiferromagnetic $FeBO_3$ by a microwave magnetic field

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A microwave magnetic field $h \cos \omega_p t$ was used in the parallel pumping configuration ($\mathbf{h} \| \mathbf{H}$) to excite transverse phonons of frequencies $\omega_k = \omega_p/2 = 2\pi\cdot(300\text{–}600)$ MHz in a sample subjected to a static magnetic field $H = 50\text{–}700$ Oe at temperatures $T = 4.2\text{–}77$ K. A size effect was observed: the parametric phonon excitation threshold decreased when the number of half-waves ($n = 140\text{–}350$) which could be fitted across the thickness of an $FeBO_3$ single crystal was an integer. A study was made of how modulation of the phonon spectrum with a field $H_m \cos \omega_m t$ influenced the amplitude of the threshold field $h_{c\|}$ under parallel pumping conditions. The experiments confirmed the validity of theoretical expressions [Eqs. (3) and (4) below] relating $h_{c\|}$ to the rate of relaxation η of the excited phonons and to the experimental parameters, and made it possible to find η by two independent methods.

INTRODUCTION

One of the most important manifestations of the spin–orbit interaction in a solid is the appearance of a coupling between elastic vibrations of the lattice and magnetic (spin) oscillations. This magnetoelastic interaction results in mixing of oscillations of different types (magnons and phonons) giving rise to new normal coupled-oscillation modes in the form of quasimagnon and quasiphonon branches. The greatest changes in the magnon and phonon spectra occur in antiferromagnets, because in these materials the relativistic magnetoelastic interaction is enhanced by the exchange interaction (for a review see, for example, Ref. 1). In the case of antiferromagnetic $FeBO_3$ (iron borate) with a magnetic easy-plane anisotropy the dispersion relation of these coupled magnetoelastic oscillations are[1,2]

$$\omega_e^2=\omega_{e0}^2+\gamma^2H_\Delta^2,$$

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

where ω_e and p_h are the frequencies of quasimagnon and quasiphonon branches; $\omega_{e0} = (\gamma^2 H(H+H_D) + v_\|^2 k_\|^2 + v_\perp^2 k_\perp^2)^{1/2}$ is the frequency of spin waves (magnons) deduced ignoring the magnetoelastic interaction; $\gamma = 2\pi\cdot 2\cdot 8$ GHz/kOe is the magnetomechanical ratio; c is the velocity of sound in the absence of any influence by the magnetic substance; k is the wave vector of quasiparticles; $\gamma H_\Delta = \gamma(H_{me}H_E)^{1/2}$ is the magnetoelastic gap in the spin-wave spectrum; H_{me} is the effective magnetoelastic interaction field; H_E is the effective exchange interaction field; $v_\|$ and $v_\perp$ are the limiting velocities of spin waves along and across the C_3 axis of the crystal; H_D is the Dzyaloshinskii field; H is the projection of the static magnetic field onto the easy plane of the crystal. In the range of wave vectors $k \lesssim 10^3\text{–}10^4$ cm^1 when the magnetic field is $H \gtrsim 100$ Oe, which is typical of our experiments, the condition $\gamma^2 H(H+H_D) \gg v^2k^2$, is satisfied and, therefore, the phonon spectrum can be regarded as linear ($\omega \propto k$) and the velocity of sound $\tilde{c}$ is a function of the magnetic field:.

$$\tilde{c}\approx c\left[\frac{H(H+H_D)}{H(H+H_D)+H_\Delta^2}\right]^{1/2}. \tag{2}$$

The coupling between the elastic and magnetic subsystems makes it possible to excite quasiphonons by an oscillatory magnetic field $h \cos \omega_p t$. The first studies of the excitation of sound in a field h in antiferromagnets were reported by Seavey[3] who studied how the frequency of the fundamental mode of transverse vibrations of thin $FeBO_3$ and α-Fe_2O_3 (hematite) plates depends on a static magnetic field. It was found that half the wavelength of sound could be fitted across the thickness of the sample; the oscillatory and static magnetic fields were applied in the easy plane of a crystal at right-angles to one another and typical frequencies of sound were $\nu \approx 10\text{–}20$ MHz. The same method was used later to excite higher harmonics of natural vibrations of a sample of $FeBO_3$ up to $n = 59$ and $\nu \approx 1$ GHz (Ref. 4). Subsequently Zhotikov and Kreĭnes in the case of $CoCO_3$ (Ref. 5) and Wettling and Jantz in the case of $FeBO_3$ (Ref. 6) demonstrated the feasibility of parametric excitation of short-wavelength phonons ($k \approx 10^4\text{–}10^5$ cm^{-1}) by the method of perpendicular transverse pumping (pump frequencies $\nu_p = 36$ GHz and $\nu_p = 9.2$ GHz, respectively). The excited phonons were recorded in Refs. 5 and 6 using the Brillouin method of light scattering. It was found that transverse phonons of frequency $\omega_{ph} = \omega_p/2$ were excited and the direction of propagation was along the C_3 axis. The law of conservation of energy and quasimomentum shows that one microwave pump photon should create a pair of phonons with approximately equal and opposite wave vectors. However, the threshold fields $h_{c\perp}$ for parametric excitation of phonons were not determined in the experiments described above. A study of perpendicular pumping in $FeBO_3$ was carried out by Kotyuzhanskiĭ and Prozorova[7] who determined the threshold field $h_{c\perp}$ as a function of the sample temperature T and the magnetic field $H(\nu_p = 35.4$ GHz) and used the values of $h_{c\perp}$ to find the functional dependence of the phonon relaxation rate η on H and T. In these estimates they used a theory of Ref. 8 relating η to $h_{c\perp}$.

In addition to perpendicular pumping, it is possible to excite phonons parametrically by the parallel pumping method ($\mathbf{h} \cos \omega_p t \| \mathbf{H}$). The threshold amplitude $h_{c\|}$ of the oscillatory fields used in parallel pumping of an easy-plane antiferromagnet was calculated in Ref. 9. Subject to the condition $\omega_p \ll \omega_c$ we find[9]

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$$\gamma_k = V_\parallel h_c, \quad \gamma_k = 2\pi\eta = 1/(2\tau),$$
$$V_\parallel = \gamma^4 H_D H_\Delta^2 \omega_{ph} / 4\omega_e^4. \tag{3}$$

Here, $V_\parallel$ is the coefficient representing the coupling of phonons to the pump field; $\omega_{ph} = \omega_p/2$; τ is the lifetime of the excited phonons.

If a static magnetic field and an oscillatory pump field are supplemented by a modulating rf field H_m such that

$$\mathbf{H} \| \mathbf{h} \cos \omega_p t \| \mathbf{H}_m \cos \omega_m t,$$

the threshold amplitude rises. A theory of this effect can be found in Refs. 10 and 11. In our case the dependence of h_c on H_m can be written as follows:.

$$\frac{h_c}{h_{c0}} - 1 = \frac{4U^2 H_m^2}{\omega_m^2 + (2\gamma_k)^2}, \quad U = V_\parallel = \frac{\partial \omega_{ph}}{\partial H}. \tag{4}$$

Here, h_{c0} is the threshold field in the absence of modulation and h_c is the field in the presence of modulation. Equation (4) is valid at moderately high modulation amplitudes ($h_c/h_{c0} < 2$) when $\omega_m > 2\gamma_k$. The increase in the critical field H_c is due to modulation of the spectrum of the excited phonons, which disturbs the average conditions for parametric resonance. A study of the influence of the modulation field on the parametric pumping threshold makes it possible to check the validity of Eqs. (3) and (4) and provides an independent method for studying the relaxation of excited oscillations.[12]

We report the first experimental investigation of parametric excitation of phonons in antiferromagnets by the parallel pumping method. We were able to achieve parametric excitation of phonons in $FeBO_3$ single crystals subjected to microwave radiation in a wide range of frequencies: $\omega_p = 2\pi\cdot(600\text{–}1200)$ MHz, $H = 100\text{–}700$ Oe. Moreover, we investigated in detail the influence of a modulating field on the parallel pumping threshold in order to check Eqs. (3) and (4). This was essential for determining whether the theory of Refs. 9–11 can be used to calculate phonon damping from the critical amplitude of an oscillatory magnetic field and from the dependences $h_c(H_m, \omega_m)$.

EXPERIMENTAL METHOD

Parametric excitation of phonons was investigated using a spectrometer working in the decimeter wavelength range.[13] Open helical resonators (with $Q \sim 500$ under load) were used as resonantly absorbing cells.

A sample of $FeBO_3$ was oriented relative to a static magnetic field H, so that H was always in the easy plane of the crystal along the easy magnetization axis of the weak ferromagnetic moment. A sample was then fitted into a Teflon holder using a bag made of cigarette paper and the whole assembly was placed inside the resonator. The resonator with the sample was filled with gaseous helium. The main measurements were carried out at $T = 77$ K. The static field was created by an electromagnet and its direction could be altered. An external modulation field H_m was applied to the helical resonator: this was done using a coil sharing the axis with the resonator. This field was measured to within 10%, but the error in the relative measurements of H_m was only 3%.

Excitation of phonons was detected under pulsed and cw conditions: it was manifested by the appearance of absorption of microwave power or by amplitude modulation, at the frequency ω_m, of the signal transmitted by the microwave resonator. Such amplitude modulation appeared immediately above the parallel pumping threshold and was associated with the excitation of forced collective oscillations in a system of parametric waves under the action of a modulation field.[11] The system used to detect the modulation response was described in detail in Ref. 13. In the absence of the modulation field the microwave signal detected in the cw regime was fed to an automatic plotter connected to a magnetic field sensor: when pulses were used, the plotter was replaced with an oscilloscope. In the pulsed regime the absorption of power due to the excitation of phonons was manifested by a characteristic distortion of the pulse profile. Since in the cw regime at high microwave oscillator powers there were effects associated with sample overheating, the main measurements were carried out by the pulse method (using pulses of 300 μs duration repeated at a frequency of 50 Hz). The amplitude of the pump field was calculated from the measured values of the input power, pump frequency, and resonator parameters. The absolute error in the determination of h_c was 30% and the relative error was 3%.

Single crystals of $FeBO_3$ had natural faces and their dimensions were $5 \times 1.7 \times 1.3$ mm or $7 \times 3 \times 1.2$ mm. The plane of growth of the plate coincided with the basal plane of the crystal.

SIZE EFFECT

Figure 1 shows a typical record obtained using our plotter, showing how the microwave power transmitted by the resonator with the sample depends on the static magnetic field when the microwave oscillator is running continuously. The magnetic field intensity was reduced from $H > H_c$ to $H = 0$. Clearly, in fields $H < H_c$ there was a considerable absorption of microwave power, corresponding to the development of a parametric instability at a fixed amplitude of microwave magnetic fields reaching the sample. The process of phonon excitation was "soft," i.e., the amplitudes of the critical field at which parametric phonons appeared and disappeared (h_{c1} and h_{c2}, respectively) agreed with the measurements within the limits of the experimental error: $h_{c1} = h_{c2} = h_c$.

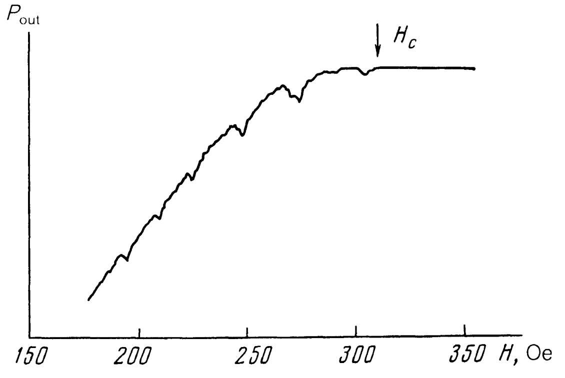

FIG. 1. Dependence of the microwave power transmitted by a resonator on the static magnetic field; $\omega_p/2\pi = 600$ MHz, $T = 77$ K. The field H_c corresponds to the onset of parametric excitation of phonons on reduction in H.

In certain magnetic fields there were some special features of the susceptibility χ'' at positions independent of the microwave power. Measurements carried out in the pulsed regime showed that an increase in χ'' was due to a reduction in the threshold field h_c at the values of H in question. The change in h_c was less than 15%. The observed features could be explained by size effects. Clearly, if the number of half-waves of the parametric phonons which can be fitted across the thickness of the sample is an integer, the Q factor of such elastic vibrations should be higher and its excitation threshold lower. The size effect was manifested for $\lambda n/2 = d$, where λ is the wavelength of the excited sound, d is the thickness of the sample, and n is an integer. In our experiments the value of n was $n = 140$–360. Allowing for the dependence of the velocity of sound $\tilde{c}$ on the magnetic field given by Eq. (2), we can write down the expression for the value of ΔH:

$$\Delta H = 2\pi c H_D H^{3/2}(H + H_\Delta^2/H_D)^{1/2}/\omega_{ph}\, d H_\Delta^2, \tag{5}$$

where ΔH is the separation between the neighboring values of the field H in which condition for the size effect is satisfied.

Figure 2 demonstrates a good agreement between the theoretical dependence of Eq. (5) and the experimental data on $\Delta H(H)$ for a sample of dimensions $7\times3\times1.2$ mm. The following were substituted into the theoretical expression: the thickness of the sample d along the C_3 axis, the velocity of transverse phonons in the same direction, and the phonon frequency $\omega_{ph} = \omega_{p/2}$. A satisfactory theoretical description of the experimental results obtained for two samples of different dimensions leads to the conclusion that transverse phonons of half the frequency propagating along the C_3 axis are created by parallel pumping. The value of H_Δ^2, which we obtained from an analysis of the size effect results, was indeed related to the dependence of the velocity of sound on the magnetic field and amounted to $H_\Delta^2 = 3.4 \pm 0.5$ kOe^2.

INVESTIGATION OF THE INFLUENCE OF THE MODULATION FIELD ON THE PARALLEL PUMPING THRESHOLD

The main measurements of the threshold amplitude of the microwave field were carried out by the pulse method.

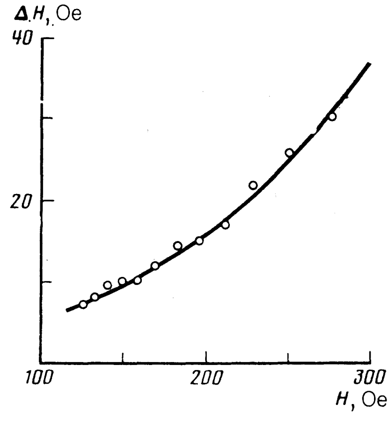

FIG. 2. Separation between adjacent features in h_c plotted as a function of a static magnetic field H for $\omega_p/2\pi = 600$ MHz at $T = 77$ K. The continuous curve is calculated from Eq. (5) using the following values of the parameters: $c = 4.7\times10^6$ cm/s (Ref. 14), $H_D = 100$ kOe (Ref. 15), $H_\Delta^2 = 3.4$ kOe^2, and $d = 0.12$ cm.

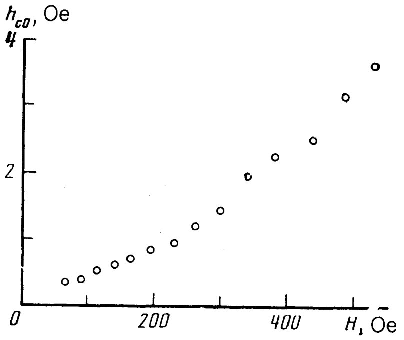

FIG. 3. Dependence of the threshold amplitude of the parallel pumping field on the static magnetic field for $\omega_p/2\pi = 1140$ MHz at $T = 77$ K.

Figure 3 shows how the threshold h_{c0} depends on the static magnetic field applied in the absence of the modulation field. In the presence of the modulation field, Eq. (4) for the pumping threshold becomes, [subject to Eq. (1)]

$$\frac{h_c}{h_{c0}} - 1 = \left(\frac{H_m}{2H}\right)^2 \left(\frac{H_\Delta^2}{HH_D + H_\Delta^2}\right)^2 \frac{\omega_{ph}^2}{\omega_m^2 + (2\gamma_k)^2}. \tag{6}$$

We can see that $(h_c/h_{c0} - 1)\alpha H_m^2, \omega_{ph}^2$. Figure 4 shows the dependence of the parameter $(h_c/h_{c0} - 1)$ on the square of the amplitude of the modulation field. The experimental points fit the linear dependence on H_m^2 well. Deviation from this linear dependence is observed at high modulation amplitudes when Eq. (6) is no longer valid.

Figure 5 shows the parameter $h_c/h_{c0} - 1$ as a function of the static magnetic field. The experimental points were obtained as follows in the range $(h_c/h_{c0} - 1) \lesssim 0.2$, where the errors were large. First, at a fixed value of H we found the dependence of the parameter $h_c/h_{c0} - 1$ on H_m. Then, the results were approximated by the function $(h_c/h_{c0} - 1)\alpha H_m^2$ (in the same way as in Fig. 4) and the point on the resultant straight line, taken at $H_m = 0.73$ Oe, was plotted (Fig. 5). This procedure made it possible to find how $h_c/h_{c0} - 1$ depends on H in a wide range of parameters. A good agreement between the experimental results and the theory was observed and in the case when $H_\Delta^2 \ll H H_D$ and

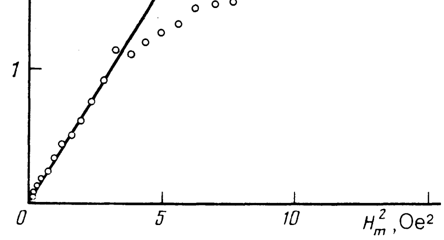

FIG. 4. Influence of the modulation field H_m on the parallel pumping threshold: $\omega_p/2\pi = 1140$ MHz, $T = 77$ K, $\nu_m = 200$ kHz, $H = 226$ Oe.

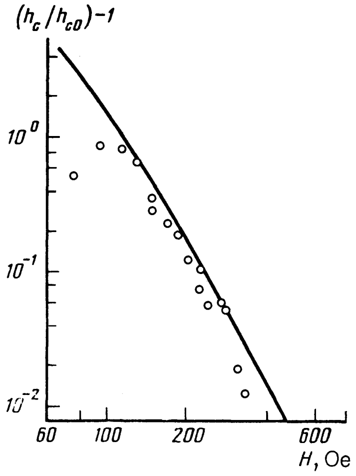

FIG. 5. Dependence of the relative increase in the pumping threshold on the static magnetic field at $T = 77$ K: $\omega_p/2\pi = 1140$ MHz, $H_m = 0.73$ Oe, $\nu_m = 300$ kHz. The curve is calculated on the basis of Eq. (6) assuming that $H_\Delta^2 = 3.4$ kOe^2. The value of 2η was calculated from the threshold field h_{c0} (Fig. 3) using Eq. (3).

$\eta =$ const the dependemce obeyed the H^{-4} law. The deviation from the theoretical dependence observed in weak fields was due to an increase in the relaxation rate η, so that the condition $v_m > 2\eta$ was no longer obeyed and Eq. (6) became invalid.

Figure 6 shows the dependence of the parameter $(h_c/h_{c0} - 1)$ on the modulation frequency obtained for different pump frequencies. The theory leading to Eq. (6) deals with the case when modulation of the natural frequency of phonons ω_k does not shift a packet in k space. In Fig. 6 this situation corresponds to the modulation frequencies to the right of the peak. However, if $\nu_m \lesssim 2\eta$, then modulation of the phonon spectrum becomes continuous so that the appearance of a parametric instability now involves groups of phonons with different wave numbers: there is an increase in the amplitude of the modes for which the geometric resonance condition is satisfied at a given moment in time and the intensities of the modes which shift out of the resonance decrease. Equation (6) then ceases to be valid.

We now consider the case $\nu_m \gtrsim 2\eta$. We represent the experimental results in terms of the dependence of $(h_c/h_{c0} - 1)^{-1}$ on ν_m^2 (Fig. 7). This dependence on the modulation frequency is in agreement with the theoretical predictions and, as demonstrated by Eq. (6), extrapolation of the linear part of the experimental curve to point of intersection with the abscissa makes it possible to find the value of the parameter (2η) . A comparison of the relaxation rate found by such extrapolation with the value of η calculated from h_{c0} using Eq. (3) is made in Fig. 8. The two methods give the same values of the relaxation parameter η when $\omega_p/2\pi = 1140$ MHz. If $\omega_p/2\pi = 603$ MHz, we can simply say that $\eta = 50$ kHz calculated from the parallel pumping threshold is within the interval $\eta = 30 \pm 30$ kHz which is deduced from an analysis of the experimental dependence $h_c(\omega_m)$ in accordance with Eq. (6).

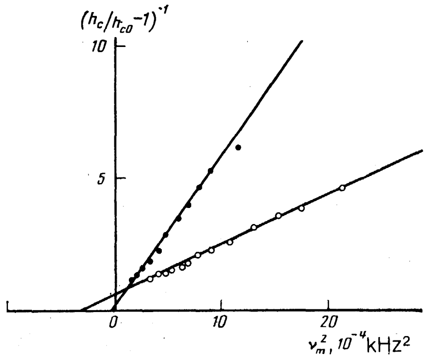

FIG. 7. Dependence of the parameter $[(h_c/h_{c0}) - 1]^{-1}$ on the square of the modulation frequency; ●) $\omega_p/2\pi = 603$ MHz, $H = 227$ Oe, $H_m = 1.57$ Oe; ○) $\omega_p/2\pi = 1140$ MHz, $H = 191$ Oe, $H_m = 1.12$ Oe. The continuous curves represent the theoretical dependence (6) with $H_\Delta^2 = 3.5$ kOe^2.

These values of η allow us to estimate the lifetime τ and the mean-free path l of phonons ($T = 77$ K, $H = 200$ Oe, $c = 4.71 \times 10^5$ cm/s):

ν_{ph}=301 MHz	τ=1.6 μs,	$l = c\tau$=0.75 cm,
ν_{ph}=570 MHz	τ=0.9 μs,	l=0.4 cm.

Therefore, the mean free path of a phonon is several times greater than the thickness of the sample (~0.1 cm), which is why we can observe the size effect.

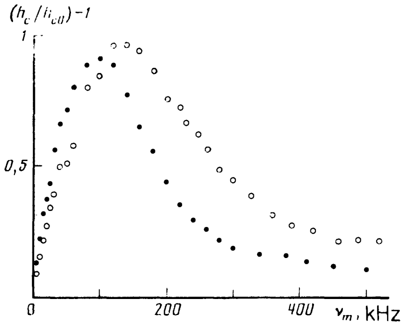

FIG. 6. Relative increase in the threshold of parallel pumping plotted as a function of the modulation frequency at $T = 77$ K: ●) $\omega_p/2\tau = 603$ MHz, $H = 227$ Oe, $H_m = 1.57$ Oe, ○) $\omega_p/2\tau = 1140$ MHz, $H = 191$ Oe, $H_m = 1.12$ Oe.

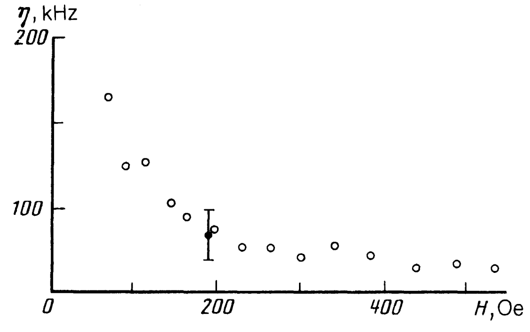

FIG. 8. Phonon relaxation rate plotted as a function of the static magnetic field ($\omega_p/2\pi = 1140$ MHz, $T = 77$ K): ●) value of η calculated from h_{c0} using Eq. (3); ○) value of η deduced from the data in Fig. 7 by the extrapolation method.

In addition to determining of η, we can use the experimental results of Fig. 7 to calculate the magnetoelastic constant H_Δ^2 which, according to Eq. (6), governs the slope of the straight lines drawn in Fig. 7. The results presented in Fig. 7 can be described by the theoretical expression (6) if $H_\Delta^2 = 3.5 \pm 0.5$ kOe^2. This value is in good agreement with H_Δ^2 deduced from the size effect.

The above analysis of the experimental data obtained at two very different pump frequencies shows that the frequency dependence of the influence of modulation on the threshold also agrees with the theoretical prediction that $h_c/h_{c0} - 1\alpha\omega_{ph}^2$.

The main result of our investigation of the influence of modulation of the phonon spectrum on the parametric excitation threshold is as follows. The theoretical expression (6) describes satisfactorily all the experimental results relating to the influence of the phonon spectrum modulation on the parallel pumping threshold. It therefore follows that the theoretical model yielding Eqs. (3), (4), and (6) is valid and we can use these equations to calculate the phonon relaxation parameter both from the critical field h_c and from the dependences of this field on the modulation field parameters.

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