Ultrasonic Absorption at Microwave Frequencies and at Low Temperatures* in MgO and Al_2O_3

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Measurements are presented of the ultrasonic absorption in MgO and Al_2O_3 for transverse and longitudinal waves in the low-temperature region. In contrast to some theoretical expectations, the absorption of longitudinal waves is not negligible but even stronger than for transverse waves. In the theoretical discussion it is shown that this strong longitudinal absorption probably is caused by three-phonon processes.

INTRODUCTION

THE interaction of ultrasonic waves with thermal phonons in dielectric crystals shows a very different behavior at high and at low temperatures. At high temperatures when the ultrasonic period $2\pi/\omega$ is much larger than the lifetime θ of most termal phonons the measured absorption can be well described by a mechanism first proposed by Akhiezer.[1–3] However, in the low-temperature region, defined by ($\omega\theta \gg 1$) experimental difficulties have prevented until recently a careful study of the hypersonic absorption, although a theory for the absorption process was already published by Landau and Rumer[4] more than 25 years ago. They pointed out that through the anharmonic cubic terms in the elastic lattice energy *transverse* acoustic waves or phonons can interact with thermal phonons in processes involving three phonons. For the absorption of *longitudinal* acoustic waves these three-phonon processes were considered unimportant on the basis of energy and momentum conservation as will be discussed later. A finite absorption was thought to result only when according to Pomeranchuk[5] fourth-order terms in the elastic energy were taken into account (so-called four-phonon processes), or when according to Herring[6,7] the elastic anisotropy of the crystal was properly considered (Herring mechanism).

In the present paper we present measurements of the absorption of transverse and longitudinal ultrasonic waves in magnesium oxide and sapphire in the low-temperature region ($\omega\theta > 1$). The interesting result is that not only the transverse but also the longitudinal absorption follows the pattern expected for a three-phonon process. The magnitude of the longitudinal absorption as well as its dependence on frequency and temperature are not compatible with a four-phonon process or the Herring mechanism mentioned above.

By these results we are led to believe that longitudinal ultrasonic waves also are able to interact strongly with thermal phonons in three-phonon processes. We have already shown in a classical calculation,[8] referred to here as paper I, that these "collinear" processes lead to a strong ultrasonic absorption, and the same result will be derived here by a quantum-mechanical argument.[8a]

This strong absorption of longitudinal waves in three-phonon processes also solves the problem of a diverging thermal conductivity which was discussed in the literature[9] under the assumption that longitudinal waves cannot be absorbed in three-phonon processes. Since they are, however, absorbed, their contribution to the heat conductivity remains finite, and no problem arises in this context.

EXPERIMENTS AND RESULTS

We have measured the attenuation of sound waves in magnesium oxide and ruby at the two frequencies of 500 and 3000 Mc/sec between 4.2 and about 100°K. The samples were single crystals of rectangular shape, 4×4×25 mm, with the 4×4-mm faces polished optically flat and parallel within a few seconds. The rod axes were oriented within one degree, parallel to the [100] axis of MgO and to the c axis of ruby. The MgO samples were colorless and quoted[10] as pure while the ruby rods contained up to 0.01% of Cr_2O_3. Care was taken to avoid resonant absorption of sound by the paramagnetic ions.[11]

For the generation and detection of the ultrasonic waves we used two methods: At 500 and at 3000 Mc/sec two x-cut quartz disks with a fundamental resonance frequency of 10 Mc/sec were bonded to the sample and excited in a high harmonic. For some experiments at 3000 Mc/sec both sides of our samples were plated with films of permalloy, 90% Ni, 10% Fe,

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[1] A. Akiezer, J. Phys. (USSR) **1**, 277 (1939).
[2] H. Bommel and K. Dransfeld, Phys. Rev. **117**, 1245 (1960).
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[4] L. Landau and G. Rumer, Z. Physik. Sowjetuuion **11**, 18 (1937).
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[7] S. Simons, Proc. Cambridge Phil. Soc. **53**, 702 (1957).
[8] R. Nava, R. Arzt, I. S. Ciccarello, and K. Dransfeld, Phys. Rev. **134**, A581 (1964).
[8a] *Note added in proof.* Arguments for this mutual interaction of parallel phonons have also recently been given by S. Simons [Proc. Phys. Soc. (London) **82**, 401 (1963)].
[9] P. G. Klemens, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1956), Vol. 14. p. 1.
[10] Supplied by R. Meller Co., Providence, Rhode Island.
[11] E. B. Tucker, Phys. Rev. Letters **6**, 183 (1961).

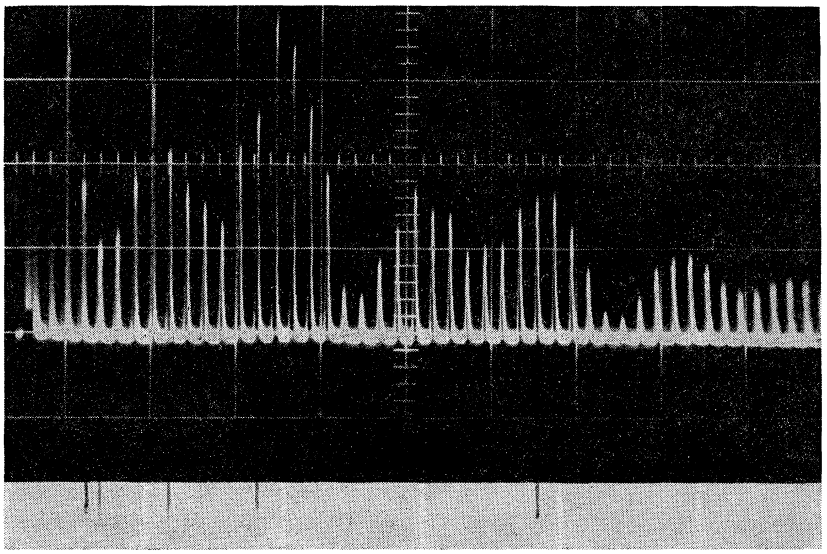


FIG. 1. Pulse pattern of 3-kMc/sec longitudinal waves in ruby at 4.2°K. The waves are excited and detected ferromagnetic resonance in thin films of permalloy (90% Ni-10% Fe) on each side of the ruby rod.

about 5000 Å thick for the generation and detection of sound by ferromagnetic resonance.[12] By using standard pulse methods sometimes more than 100 acoustic echo pulses were received.

Figure 1 shows a typical pulse pattern, the time between two consecutive pulses corresponds to the time for one round trip in the rod. The decay of the pulse pattern is hardly ever exponential. The reasons for the irregular amplitudes of the various pulses are several: the end faces may not be sufficiently parallel, the rod may not be elastically homogeneous, etc., all of these effects being temperature-independent. The absorption at a temperature T was determined from the decrease of an echo pulse relative to its magnitude at 4.2°K.

With ferromagnetic excitation we occasionally observed a drastic change in the relative amplitude of the peaks on varying the intensity or direction of the magnetic field. We believe that this is caused by interference effects which arise from mechanical or magnetic nonuniformities of the films, or the excitation of Walker modes. By improving our deposition methods and by using films of small cross section these phenomena could be avoided.

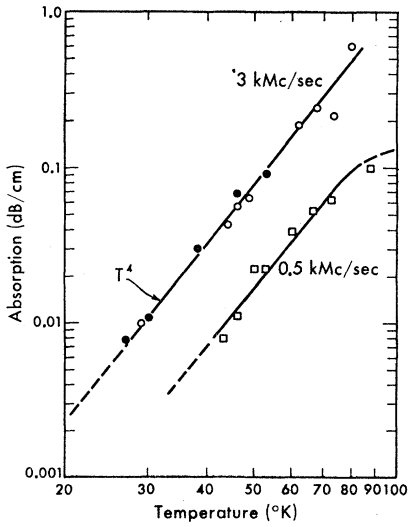


FIG. 2. Attenuation of longitudinal waves in ruby as a function of temperature.

Through our long ruby rod only longitudinal waves could be transmitted well. The propagation of transverse waves proved to be considerably more difficult. In general, only one transverse echo could be seen. This difficulty probably arise from the acoustical birefringence, so that along the c axis the acoustic wave vector and Poynting vector are not parallel, and the acoustic energy does not travel parallel to the rod axis. The absorption of longitudinal waves versus temperature is plotted in Fig. 2. Several runs at 3000 Mc/sec, with different ruby rods carrying different magnetic films gave the reproducible results of Fig. 2: The absorption increases with temperature approximately as T^4. For the investigation of the frequency dependence, measurements at 500 Mc/sec, which agree with the results of Fitzgerald and Truell,[13] are also included in Fig. 2; the frequency dependence is close to linear.

The absorption of longitudinal and transverse waves in MgO is plotted versus temperature in Figs. 3(a) and 3(b). The data for longitudinal waves, although not as accurate as the transverse data, clearly indicate that the frequency dependence is again close to linear, and the temperature dependence not far from T^4.

With longitudinal waves the acoustic power level was kept low enough to avoid the nonlinear effects reported by Shiren,[14] and for transverse waves such a power-dependent absorption could not be observed even

[12] H. Bommel and K. Dransfeld, Phys. Rev. Letters **3**, 83 (1959).

[13] T. M. Fitzgerald and R. Truell, research report, Brown University, 1963 (unpublished).

[14] N. S. Shiren, Phys. Rev. Letters **11**, 3 (1963).

at considerably higher power levels than those used by Shiren. To our knowledge, for shear waves in the kMc/sec range, these are the first data on the temperature dependence in a material other than quartz.

Both for transverse and longitudinal waves in MgO a noticeable additional absorption occurred around 15°K, when the absorption was measured in zero magnetic field. Figure 3(a) also shows this extra absorption at 3000 Mc/sec, which could be removed by applying a magnetic field of the order of 1000 Oe. A similar phenomenon was observed by Bolef *et al.*[15] at lower frequency and lower temperature in a more drastic way in heavily doped MgO. More experiments are planned for a detailed investigation of this anomaly which is probably caused by paramagnetic impurities.

DISCUSSION

Our experiment results may be summarized by saying that at low temperatures for both transverse and longitudinal waves the absorption increases linearly with frequency and with the fourth power of the temperature. In perfect crystals the absorption is due only to scattering by other thermal phonons, and is possible only if the potential energy developed in powers of the strain contains cubic or higher order terms. Landau and Rumer[4] considering the cubic terms only calculated the absorption of transverse waves for low temperatures, when $\omega\theta\gg 1$. They found

$$\alpha_t \approx \tfrac{1}{4}(2Q+6R)^2(1-V_t/V_L)^2 \frac{k^4}{\rho^3 V_L{}^{10}\hbar^3}\omega T^4, \qquad (1)$$

where Q and R are constants of the order of 5×10^{12} for ruby and MgO, k is the Boltzmann constant, and ρ the density. The agreement with our results on MgO is satisfactory. The absorption is of the right order of magnitude and varies with temperature as T^4, as expected. Expression (1) was derived only for *transverse* waves, with the observation that the conservation of energy and momentum rules out three-phonon processes for the absorption of *longitudinal* waves.

Pomeranchuk[5] invoked the much smaller fourth-order terms of the elastic energy to account for the scattering of longitudinal phonons. The absorption according to his calculations should be proportional to ω^2T^7, which is in strong disagreement with our experiments, both for ruby and MgO. In addition, Orbach[16] has already estimated, for the case of quartz, that Pomeranchuk's calculations give an absorption which is several orders of magnitude smaller than the observations. We may therefore safely conclude that a four-phonon process is not responsible for the observed absorption of longitudinal waves.

On the other hand, the Herring mechanism[6] also cannot account for our results. The absorption due to this process was calculated by Simons[7] for cubic crystals. For a longitudinal wave propagating parallel to the (100) axis he found

$$\alpha = A\omega^2T^3. \qquad (2)$$

The quadratic frequency dependence is not compatible with our observations. The factor A depends on the crystal and can be estimated for MgO, with the result that for our frequencies and temperatures the absorption according to (2) is about three orders of magnitude smaller than the measured values. For ruby the absorption by the Herring mechanism has not been calculated numerically, but should be proportional to ω^3T^2, which agrees even less with our experiments.

In view of the fact that our data with longitudinal waves behave as if they are caused by three-phonon processes, and in view of the difficulty in explaining them in any other way, we proceed to re-examine in more detail the absorption of longitudinal waves by three-phonon processes. We have already shown classically in paper I that in isotropic solids the absorption of longitudinal waves by three-phonon processes is of similar magnitude as the absorption of transverse waves. But the fact that a longitudinal acoustic wave can interact in three-phonon processes with almost as many thermal phonons as a transverse wave may be shown in a simpler argument, which will be presented here:

The conservation of energy and momentum requires

$$\omega_1+\omega_2=\omega_3, \qquad (3)$$

$$\mathbf{k}_1+\mathbf{k}_2=\mathbf{k}_3, \qquad (4)$$

where the index "1" refers to the ultrasonic phonon. Since in our temperature range $|k_1|\ll|k_{2,3}|$ the momentum conservation (4) can be expressed in terms of the angle α between the acoustic wave vector $\mathbf{k}_1$ and $\mathbf{k}_2$ (see Fig. 4).

$$\cos\alpha=(|k_3|-|k_2|)/|k_1|,$$

or, because of

$$|k_1|=\omega_1/v_1, \quad |k_{2,3}|=\omega_{2,3}/v_{\rm ph},$$

we can write for (4)

$$\cos\alpha=\frac{v_1}{v_{\rm ph}}\left(\frac{\omega_3-\omega_2}{\omega_1}\right), \qquad (5)$$

where α is the angle under which a thermal phonon with velocity $v_{\rm ph}$ can interact with an acoustic wave of velocity v_1 with conservation of energy and momentum.

But if energy is not exactly conserved because of the uncertainty in the energy arising from a finite relaxation time of θ of the thermal phonons, we find

$$\omega_3-\omega_2-\omega_1\leqq 1/\theta$$

and $\cos\alpha$, according to (5), can assume any value between the two limits

$$\cos\alpha_{\max}=\frac{v_1}{v_{\rm ph}}\left(1-\frac{1}{\omega_1\theta}\right); \quad \cos\alpha_{\min}=\frac{v_1}{v_{\rm ph}}\left(1+\frac{1}{\omega_1\theta}\right). \qquad (6)$$

[15] D. I. Bolef, J. DeKlerk, and R. B. Gosser, Rev. Sci. Instr. 33, 631 (1962).

[16] R. L. Orbach, thesis, University of California, Berkeley, 1960 (unpublished).

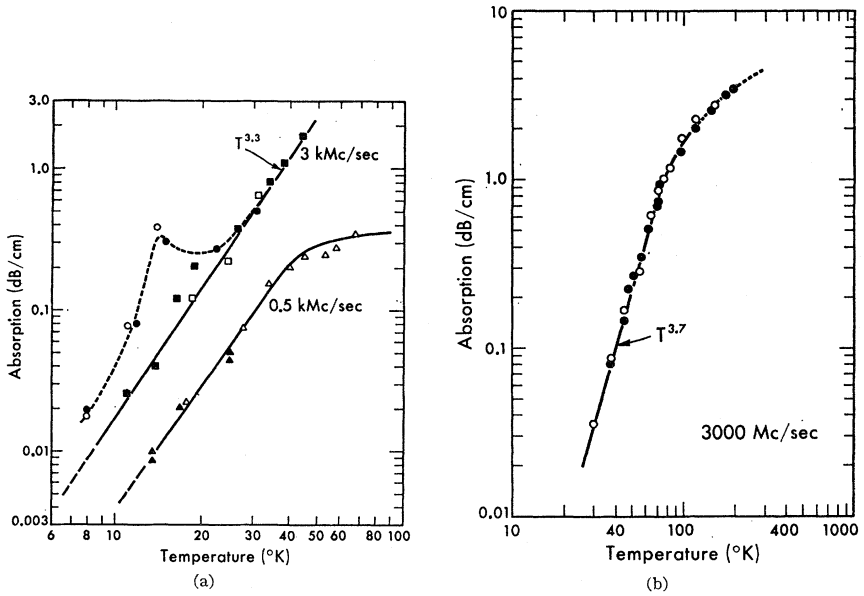


FIG. 3. Attenuation of ultrasonic waves in MgO as a function of temperature. The dashed curve gives the attenuation in zero external magnetic field; the continuous ones in a field of about 4500 Oe. (a) Longitudinal waves, (b) transverse waves.

Clearly, because of the uncertainty in energy, a thermal phonon can interact with an acoustic phonon between the angles $\alpha_{\max}$ and $\alpha_{\min}$ without violating the conservation of energy and momentum. For *transverse* waves, for example, we find from (6)

$$\alpha_{\max}-\alpha_{\min}=\Delta\alpha=\frac{2\cos\alpha}{\sin\alpha}\frac{1}{\omega_1\theta}. \tag{7}$$

This corresponds to a relative solid angle

$$\frac{\Delta N}{N}=\frac{2\pi\sin\alpha}{4\pi}\frac{2\cos\alpha}{\sin\alpha}\frac{1}{\omega_1\theta}=(\cos\alpha)\frac{1}{\omega_1\theta}. \tag{8}$$

For *longitudinal* waves ($v_1=v_{\rm ph}$, $\alpha_{\min}=0$, collinear case) we find from (6)

$$\tfrac{1}{2}(\Delta\alpha)^2=1/\omega_1\theta,$$

and for the relative solid angle

$$\frac{\Delta N}{N}=\frac{\pi\alpha_{\max}{}^2}{4\pi}=\frac{1}{2}\left(\frac{1}{\omega_1\theta}\right). \tag{9}$$

On comparing (8) and (9) it is evident that the two solid angles are almost identical for any relaxation time θ, as long as $\omega_1\theta>1$, and the dispersion is neglected.

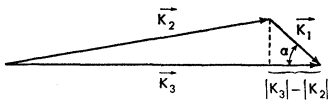


FIG. 4. Momentum conservation for the absorption of an acoustic phonon with wave vector $\mathbf{k}_1$ by a thermal phonon of wave vector $\mathbf{k}_2$. $\mathbf{k}_3$ is the wave vector of the final phonon in this three-phonon process.

One should therefore expect that a quantum-mechanical treatment of the ultrasonic absorption due to three-phonon processes should give very similar results for transverse and longitudinal waves. We made such a calculation, following the method of Landau and Rumer,[4] and we found for the absorption of longitudinal waves due to the interaction with collinear thermal phonons:

$$\alpha_L\approx\tfrac{1}{2}[6P]^2\frac{k^4}{\rho^3 V_L{}^{10}\hbar^3}\omega T^4. \tag{10}$$

P is a constant of the same order of magnitude as Q and R in Eq. (1), and therefore the absorption of longitudinal waves is not too different from the absorption of transverse waves.

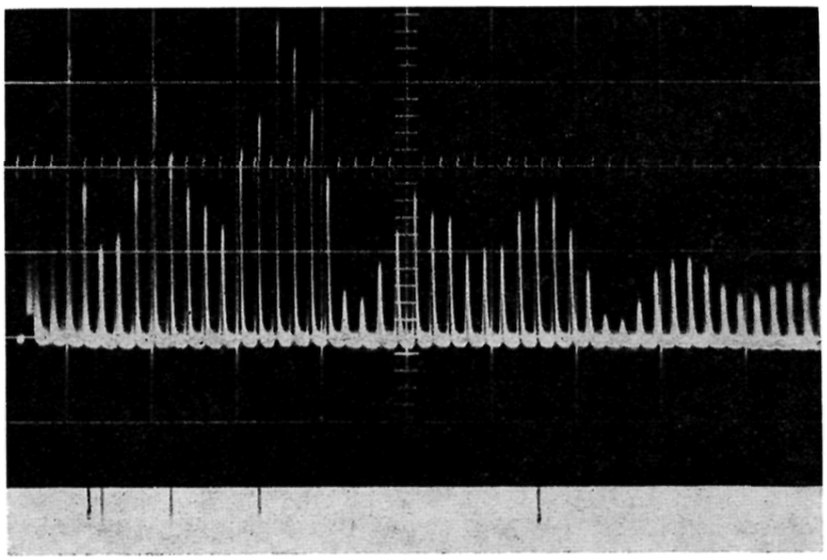
In his argument we have neglected the dispersion. As shown in paper I this neglect is justified as long as

$$\omega\theta\leqq 2\pi(T_D/T)^2,$$

with T_D being the Debye temperature of the crystal. For our samples below 60°K $2\pi(T_D/T)^2\approx 1500$ and for our frequencies $\omega\theta$ is indeed smaller than 1500, if the mean free path of the thermal phonons remains below 1 mm. Thus, the dispersion can be neglected.

It seems to be a characteristic feature of cubic crystals that the absorption of longitudinal waves is noticeably stronger than for transverse waves. This is evident from our measurements on MgO and was also found in Ge by Dobbs *et al.*[17]

[17] E. R. Dobbs, B. B. Chick, and R. Truell, Phys. Rev. Letters 3, 332 (1959).



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