## Ultrasound absorption in solids at helium temperatures

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Absorption of ultrasound in a number of substances is studied in a broad frequency range at helium temperatures. The available published data and the results obtained in the present work are systematized. A correlation between ultrasound absorption and the Debye temperature is observed at helium temperatures. A hypothesis regarding the nature of the "residual" losses is advanced.

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At helium temperatures, all the known crystals satisfy, even at frequencies  $f \ge 0.1$  GHz, the condition

$$\omega \tau_{\rm ph} > 1, \tag{1}$$

where  $\omega$  is the ultrasound frequency and  $\tau_{ph}$  is the average lifetime of the thermal phonons, which can be estimated from measurements of the thermal conductivity K:

$$\tau_{\rm ph} = \frac{3K}{C_{\rm v} \langle v \rangle^2} \tag{2}$$

 $(C_v$  is the heat capacity of the medium and  $\langle v \rangle$  is the average thermal velocity). Satisfaction of condition (1) corresponds to the mechanism developed in the classical paper of Landau and Rumer<sup>[1]</sup> for the absorption of a sound wave by thermal phonons. The expression for the damping is

 $\alpha_{LR} \sim \omega T^{*} \tag{3}$ 

## (T is the crystal lattice temperature).

The Landau-Rumer absorption of sound waves was investigated experimentally in very great detail for a large number of solids. The results of these investigations are contained in a number or reviews.<sup>[2,3]</sup> In essence, one refinement must be introduced in these studies. When plotting the temperature dependence of the absorption, the authors usually observe the temperature-independent absorption section in the interval of T from 4.2 to 20-40 K. This section of the curve is called the "residual" losses. The quantity compared with the theory of Landau and Rumer<sup>[1]</sup> or with its modifications<sup>[4]</sup> is  $\alpha - \alpha_{res}$ , and a good qualitative agreement is obtained with the calculation. The residual losses,  $\alpha_{res}$ , are usually attributed to imperfections of the crystal structure, of the finish of its acoustic planes, etc.

Abundant material on the residual losses has by now been accumulated for a large group of crystals. Thus, the anisotropy of the absorption  $\alpha_{res}$ , for example for LiF, <sup>[5]</sup> has revealed a frequency dependence. <sup>[6,7]</sup> It is somewhat surprising that in the most perfect and purest crystals of the semiconductors InSb and Ge<sup>[6,8]</sup> these losses are larger by two or three orders of magnitude than in Al<sub>2</sub>O<sub>3</sub><sup>[9]</sup> or  $\beta$ -B, <sup>[10]</sup> which were synthesized at a high temperature ~ 2000 K and had a large number of structure imperfections. Moreover, direct experiments in which a large number of impurities has been introduced into the crystals GGG<sup>[11]</sup> and TiO<sub>2</sub><sup>[7]</sup> (up to  $10^{20} \text{ cm}^{-3}$ ) did not influence, within the limits of experimental error, the value of  $\alpha_{\text{res}}$ . The foregoing data justify, in our opinion, a review of the traditional concepts concerning the nature of the residual losses.

We present here a number of new experimental data on the values of  $\alpha_{\rm res}$  in a wide range of frequencies for a number of materials, and systematize the known published data.

In the experimental measurements of the residual absorption, particular significance attaches to the methodological problems connected with elimination of the traditional error sources, such as roughness of the surface, non-parallelism of the acoustic mirrors, etc. In the InSb, Ge, and GaAs crystals investigated by us the concentration of the uncontrollable impurities did not exceed  $10^{16}$  cm<sup>3</sup>, and the dislocation density did not exceed  $10^4$  cm<sup>-2</sup>. The LiNbO<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub> crystals were of high optical and acoustical quality. In a number of points the values of  $\alpha_{\rm res}$  measured by us for these crystals agreed with the known published data. Measurement of  $\alpha_{res}$  was carried out by the well-known echo method, with the sound excited by CdS converters or by surface excitation of LiNbO<sub>3</sub>. (The typical samples measured 0.2×0.2×1.5 cm). The acoustic mirrors (end planes of the samples) were polished with class-14 accuracy and the quality of the polish was monitored by optical methods. The parallelism of the mirrors, monitored with a GS5 goniometer, was not worse than 4". The nonparallelism of the acoustic mirrors was taken into account in the measurements of  $\alpha_{res}$ at frequencies 5-9 GHz. The greater part of the measurements was performed at frequencies  $f \ge 0.5$  GHz. This choice of the frequencies range makes it possible to reduce to a minimum the diffraction losses connected with the divergence of the sound beam; these losses are proportional to  $\lambda_{ac}/d$  (d is the characteristic dimension of the acoustic radiator).

The values of  $\alpha_{\rm res}$  for a large group of substances were systematized at the value for 1 GHz. The reason is that most measurements reported in the literature were performed at this frequency or frequencies very close to it. In addition, as noted above, the diffraction losses are not so significant at this frequency and the deviation of the acoustic mirrors of the sample from parallelism still does not come into play, provided that it does not exceed 5 to 10". The errors in the determination of the coefficient of the lattice absorption of the ultrasound did not exceed, according to our esti-



FIG. 1. Frequency dependences of the residual absorption of longitudinal waves in InSb, Ge,  $Al_2O_3$ , and of shear waves in LiNbO<sub>3</sub>. Dark symbols—data obtained by others for InSb, Ge, <sup>[8]</sup> and LiNbO<sub>3</sub>. <sup>[20]</sup>

mates, 0.1-0.5 dB/cm (depending on the absolute value of the absorption).

Figure 1 shows a series of plots of the residual sound absorption against the frequency for a number of substances. At relatively low frequencies, the dependence of  $\alpha_{\rm res}$  on the frequency is close to quadratic, and at higher frequencies the curves have a tendency to flatten out. If it is assumed that sound absorption is a relaxation process, then the observed experimental dependence can be described by the expression

$$\alpha_{\rm res} \sim \frac{\omega^2 \tau_{\rm rel}^2}{1 + \omega^2 \tau_{\rm rel}^2},\tag{4}$$

where  $\tau_{re1}$  is the characteristic time of relaxation of the perturbation due to the sound wave to equilibrium. Values  $10^{-11}$  sec are obtained for  $\tau_{re1}$ . We confine ourselves to this rough order-of-magnitude estimate, although it is seen that this time is different in different



FIG. 2. Dependence of the residual absorption on the Debye temperature at 1 GHz in InSb.<sup>[6]</sup> InAs,<sup>[12]</sup> GaSb.<sup>[13]</sup> Ge,<sup>[8]</sup> GaAs,<sup>[16]</sup> LiNbO<sub>2</sub>,<sup>[20]</sup> Si.<sup>[24, 26]</sup> TiO<sub>2</sub>,<sup>[7]</sup> MgO,<sup>[15]</sup> LiF<sup>[5]</sup>, CaWO<sub>4</sub>,<sup>[18]</sup> SiO<sub>2</sub>,<sup>[17]</sup> GGG,<sup>[11]</sup> YAG,<sup>[19]</sup> Al<sub>2</sub>O<sub>3</sub>,<sup>[9]</sup>  $\beta - B$ .<sup>[10]</sup> The data for GaAs and Si were obtained by extrapolation to 1 GHz.  $\circ$ -absorption of longitudinal waves,  $\bullet$ -absorption of shear waves. The straggling of the available absorption data for waves of different polarizations, propagating in different directions, is indicated.



FIG. 3. Temperature dependences of the absorption of longitudinal sound waves at 1 GHz in  $InSb^{[6]}$  and in  $\beta$ -B.<sup>[10]</sup>

materials. We note only that  $\tau_{rel} \ll \tau_{yh}$ .

One cannot fail to note the large difference between the values of  $\alpha_{\rm res}$  for longitudinal and shear waves. In all the investigated cases  $\alpha_{\rm res}^{\rm long} < \alpha_{\rm res}^{\rm shear}$ . The difference in the absolute values reaches a factor of two to three. This illustrated by the data on the crystals InSb<sup>[6]</sup>, LiF<sup>[7]</sup>, Al<sub>2</sub>O<sub>3</sub><sup>[9]</sup>, LiNbO<sub>3</sub> and others. The residual absorption is strongly anisotropic (LiF, TiO<sub>2</sub>). These experimental results cannot be interpreted as being due to the known mechanisms of sound absorption at helium temperatures (dislocations, impurities, the presence of boundaries, etc.).

The singularities noted above suggest that the residual losses are determined not by structural or other defects of the investigated samples, but by some fundamental physical properties. A generalized albeit sufficiently approximate characteristic of the elastic oscillations of the solid is the Debye temperature  $\Theta$ . We have attempted to systematize our own and the known published values of  $\alpha_{\rm res}$  as functions of the Debye temperature (Fig. 2). The values of  $\alpha_{\rm res}$  were taken at T = 4.2 °K and f = 1 GHz; when data are available, values are given for the longitudinal and shear waves. In spite of the appreciable scatter, a dependence of  $\alpha_{\rm res}$ on  $\Theta$  can be discerned, approximately described by the expression

$$\alpha_{\rm res} \sim \Theta^{-n}, \quad n = 3 \pm 1. \tag{5}$$

A similar dependence of  $\alpha$  on  $\Theta$  is typical of the region  $\omega \tau_{ph} \ll 1$ , when an external sound wave interacts with the entire ensemble of thermal phonons, and the absorption has a relaxation character and correlates with the Debye temperature. <sup>[21,25]</sup> A correlation is observed between the temperature interval in which  $\alpha = \text{const}$  (Fig. 3) and the Debye temperature. The point at which the absorption begins to depend on the temperature is determined with a considerable degree of error, but nevertheless one can observe a tendency of the temperature interval in which  $\alpha$  is constant to broaden with increasing Debye temperature (Fig. 4).

The material presented above is purely experimental and its final interpretation is not yet clear. We shall dwell briefly on the traditional methods of explaining the sound absorption at low temperatures, similar to the methods analyzed by Holland.<sup>[22]</sup> Absorption by impurities and isotopic impurities is proportional to  $\omega^4$ 



FIG. 4. The temperature corresponding to the start of the section of the temperature-independent excitation, as a function of the Debye temperature.

and its absolute magnitude is  $\approx 10^{-9}$  dB/cm which does not agree with all the known experimental data. Absorption by dislocation is proportional to  $\omega$ , but the absolute value is estimated at  $10^{-2}-10^{-3}$  dB/cm (at a frequency 1 GHz) at  $N_d = 10^6$  cm<sup>-2</sup>.

Thus, the residual absorption can be explained only by invoking a new physical mechanism, possibly the interaction with the zero-point lattice vibrations. Their energy is  $E_0 \approx Nk\Theta$ , where N is the number of atoms per unit volume and k is Boltzmann's constant; this energy depends little on the temperature and contains the entire spectrum of the crystal-lattice vibrations. Modulation of the energy of the zero-point vibrations can lead to absorption of the sound wave. This process is usually neglected.<sup>[22]</sup> The proposed hypothesis calls for rigorous theoretical calculations.

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<sup>1</sup>L. D. Landau and Yu. B. Rumer, Zh. Eksp. Teor. Fiz. 11, 18 (1937); L. D. Landau, Sobranie trudov (Collected Works), No. 1, Nauka, 1965, p. 227.

- <sup>2</sup>V. V. Lemanov and G. A. Smolenskii, Usp. Fiz. Nauk 108, 465 (1972) [Sov. Phys. Usp. 15, 708 (1973)].
- <sup>3</sup>R. Truell *et al.*, Ultrasonic Methods in Solid State Physics, Academic, 1969.
- <sup>4</sup>H. J. Maris, Philos. Mag. 9, 901 (1964); Thesis, Imperial College, London, 1963.
- <sup>5</sup>R. C. Hanson, J. Phys. Chem. Solids 28, 475 (1967).
- <sup>6</sup>S. N. Ivanov, G. D. Mansfeld, and E. N. Khazanov, Fiz. Tverd. Tela 15, 2972 (1973) [Sov. Phys. Solid State 15, 1982 (1974)]; S. N. Ivanov, I. M. Kotelyanskii, and E. N.
- Khazanov, Fiz. Tverd. Tela 17, 349 (1975) [Sov. Phys. Solid State 17, 220 (1975)].
- <sup>7</sup>J. N. Lange, Phys. Rev. 176, 1030 (1968); Phys. Rev. 159, 703 (1967).
- <sup>8</sup>E. Groubert and A. Boyer, Phys. Status Solidi 5, 499 (1971).
- <sup>9</sup>J. de Klerk, Phys. Rev. **1**39 [A] 1635 (1965).
- <sup>10</sup>J. D. Young, D. W. Oliver, and G. A. Slack, Appl. Phys. Lett. 14, 301 (1969).
- <sup>11</sup>M. Duilot, J. Appl. Phys. 45, 2836 (1974).
- <sup>12</sup>M. J. Keck and R. J. Sladec, Phys. Rev. 185, 1083 (1969); Phys. Rev. [B] 2, 3135 (1970).
- <sup>13</sup>E. Croubert and A. Boyer, Intern. Conf. on Phonon Scattering in Solids, Paris, 1972, p. 150.
- <sup>14</sup>Yu. V. Gulyaev, A. M. Kmita, and A. V. Medved', Fiz. Tverd. Tela 12, 690 (1970) [Sov. Phys. Solid State 12, 536 (1970)].
- <sup>15</sup>J. de Klerk, Final Report AFCRL 64-904 on Contract A. F. 19 (604)-8826, 1964.
- <sup>16</sup>R. I. Cottan and G. A. Saunders, J. Phys. C 7, 2447 (1974).
- <sup>17</sup>J. de Klerk, Ultrasonics 2, 137, July-Sept., 1964.
- <sup>18</sup>M. F. Lewis, Phys. Lett. 38, 465 (1972).
- $^{19}\mathrm{M.}$  E. Lewis and E. Patterson, J. Appl. Phys. 44, 10 (1973).
- <sup>20</sup>E. C. Spenser and P. V. Lenzo, J. Appl. Phys. 38, 423 (1967); R. W. Kedzic, M. Kestigiane, and A. B. Smith, Appl. Phys. Lett. 9, 155 (1966).
- <sup>21</sup>A. I. Akhiezer, Zh. Tekh. Fiz. 8, 1318 (1938).
- <sup>22</sup>M. C. Holland, IEEE Trans. Sonics and Ultras. SU-15, 18 (1968).
- <sup>23</sup>W. Maseon, ed. Physical Acoustics, Vol. 3, Academic, 1966. [Russian transl., Mir, 1968, p. 311].
- <sup>24</sup>M. Pomerantz, Proc. IEEE 53, 1624 (1965).
- <sup>25</sup>D. W. Oliver and C. A. Slack, J. Appl. Phys. 37, 1542 (1966).
- <sup>26</sup>A. Zemel and Y. Goldstein, Phys. Rev. [B] 7, 191 (1973).

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