INVESTIGATION OF THE PHONON HEAT CONDUCTION OF MONOCRYSTALS BY A QUANTUM ACOUSTIC METHOD

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A comparative investigation is made of the heat conduction of ruby, quartz, and lithium niobate by direct measurement and by a calculation using the Debye formula on the basis of phonon interaction data from measurements of the absorption of hypersound. Methods are described and results of measurements of the heat conductivity, ultrasound velocity, and hypersound absorption coefficients are obtained in a broad temperature range.

The investigation of the heat conductivity of monocrystals on the basis of microscopic representations proposes taking account of various phonon—phonon interactions which result in finiteness of the phonon lifetime. Namely this phonon lifetime indeed determines the magnitude of the coefficient of heat conductivity of an isotropic solid according to the classical Debye formula [1]

$$\lambda = \frac{1}{3} C_V V_p^2 \bar{\tau}_p. \tag{1}$$

The phonon velocity is hence connected with the velocities of longitudinal and transverse sound in the sample under investigation by the relationship

$$\frac{3}{V_p^3} = \frac{1}{V_l^3} + \frac{2}{V_t^3}. \tag{2}$$

A strict quantum mechanical computation of the time $\bar{\tau}_p$ for specific crystals is fraught with substantial computational difficulty (see [2], say). Hence, it is necessary to obtain information about the behavior and magnitude of $\bar{\tau}_p$ (in addition to the well-known, but not always accessible, method of inelastic incoherent scattering of cold neutrons) from independent measurements.

Finding the phonon lifetime in real crystals from measurements of the sound absorption coefficients in diverse temperature bands is considered in this paper. It is interesting to note that the possibility, in principle, of such a relationship was mentioned by Debye in his already mentioned classical paper [1]. However, only the subsequent construction of a quantum theory of solids permitted establishment of this relationship successively and strictly taking into account of the whole set of sound wave interaction processes with the vibrating crystal lattice. Thus, considering just the interaction with longitudinal vibrations of the type L + L → L as basic, we obtain the following expression for the absorption coefficient α of a longitudinal sound wave of frequency ω at a temperature T (see [3] the relationships (62.27), (K43), (K45)):

$$\alpha(\omega, T) = \omega \int d\omega_k f(T, \omega_k) \left\{ \operatorname{arctg}[2\omega\tau_k(\omega_k)] - \operatorname{arctg}\left[\frac{1}{8}\left(\frac{T}{\theta_D}\right)^2 \omega\tau_p(\omega_k)\right]\right\}, \tag{3}$$

$$f(T, \omega_k) = \frac{A^2(\omega_k)\hbar}{8\pi^2\rho^3 V_l^{10}} \omega_k^2 \left(\frac{\partial N}{\partial \omega_k}\right). \tag{4}$$

Averaging the vibrations of different polarization over the true phonon distribution $N(\omega_k)$ is the main

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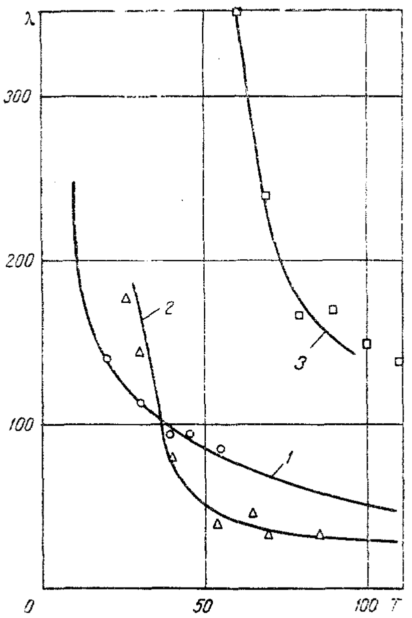

Fig. 1

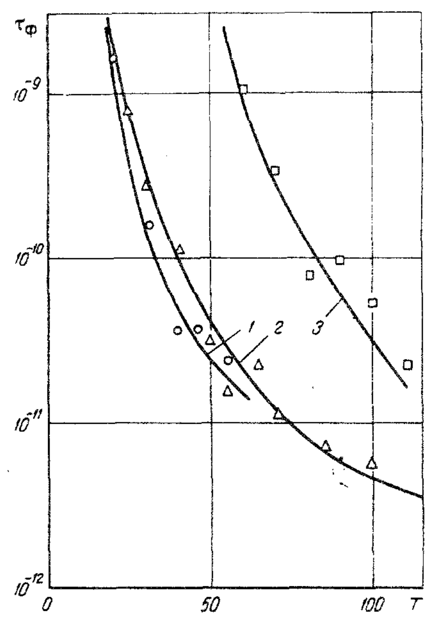

Fig. 2

Fig. 1. Dependences of the coefficient of heat conductivity λ (W/M · °K) on the temperature T (°K) for ruby (Al_2O_3) — 3, quartz (SiO_2) — 1, and lithium niobate ($LiNbO_3$) — 2. The solid lines denote λ measured by the Fourier method ($Q = \lambda S \,\mathrm{grad}\, T$) and the points denote the λ computed by the Debye formula ($\lambda = 1/3 C_V V_p^2 \tau_p$).

Fig. 2. Dependence of the mean phonon lifetime τ (sec) on the temperature T (°K) for ruby (Al_2O_3) — 3, quartz (SiO_2) — 1, and lithium niobate ($LiNbO_3$) — 2. The solid lines denote τ_p obtained from thermal measurements, and the points denote the τ_p computed from the coefficients of hypersound absorption (1)-(9).

difficulty in the direct evaluation of this quantity. However, the ratio between the sound absorption coefficients for two different frequencies can be taken to find the mean phonon lifetime $\bar{\tau}_p$ for a given temperature. We obtain the following simple expression:

$$\frac{\alpha_1}{\alpha_2} \equiv \frac{\alpha(\omega_1, T)}{\alpha(\omega_2, T)} = \frac{\omega_1}{\omega_2} \cdot \frac{\mathrm{arctg}\left\{\dfrac{(2-\gamma)(\omega_1\bar{\tau}_p)}{1+2\gamma(\omega_1\bar{\tau}_p)}\right\}}{\mathrm{arctg}\left\{\dfrac{(2-\gamma)(\omega_2\bar{\tau}_p)}{1+2\gamma(\omega_2\bar{\tau}_p)}\right\}}, \tag{5}$$

where we have used the notation

$$\gamma = 4{,}87\left(\frac{T}{\theta_D}\right)^2. \tag{6}$$

The form of (5) shows that all the quantities dependent on the force constants of the crystal and on the true shape of its phonon spectrum and governing the explicity form of $\alpha(\omega, T)$ are canceled in this case.

We find directly from (5) in diverse temperature and frequency ranges of the sound being absorbed:

$$\bar{\tau}_p^2 = \frac{3}{4} \cdot \frac{(\alpha_1\omega_2^2 - \alpha_2\omega_1^2)}{(\alpha_1\omega_2^4 - \alpha_2\omega_1^4)}, \quad (\omega\bar{\tau}_p) < 1; \tag{7}$$

$$\bar{\tau}_p = \frac{1}{\pi} \cdot \frac{(\alpha_1 - \alpha_2)}{(\alpha_1\omega_2 - \alpha_2\omega_1)}, \quad 1 < (\omega\bar{\tau}_p) < \frac{1}{\sqrt{\gamma}}; \tag{8}$$

$$\tau_p^2 = \frac{(\alpha_2\omega_2^2 - \alpha_1\omega_1^2)}{3\gamma^2(\omega_1\omega_2)^2(\alpha_2 - \alpha_1)} \quad (\omega\tau_p) \gg 1 \tag{9}$$

The relationships (7)-(9) were used to investigate the heat conductivity and sound absorption in ruby, quartz, and lithium niobate monocrystals.

TABLE 1. Dependence of the Specific Heat C_p (J/kg, °K) of Ruby (Al_2O_3), Quartz (SiO_2), and Lithium Niobate ($LiNbO_3$) on the Temperature (°K)

T, °K	20	30	40	60	80	100	200
Al_2O_3	2	7	13	45	98	145	684
SiO_2	42	70	90	140	180	230	460
$LiNbO_3$	4	9	26	80	160	250	624

The Liss method [4] with some structural changes associated with the need to conduct the measurements in a broader temperature range was used to measure the heat conductivity. The heat conductivity of monocrystals was measured at temperatures from 5-300°K. At low temperatures the side heat losses from the specimen could be neglected, hence, the Fourier formula was used in the following form to calculate the heat conductivity:

$$\lambda = \frac{Q\Delta x}{S\Delta T}\,. \tag{10}$$

The specimens were monocrystals, cut along the optical axis for ruby and lithium niobate and along the piezoelectric axis for quartz, in the form of rods 15-30 mm long and 3 mm in diameter. All the measurement results were recorded on the tape of a six-point ÉPP-09 potentiometer, for which dc amplifiers of F16 type were used to increase the response. Experience showed that it is simpler to process measurement results recorded on the potentiometer tape, and they are obtained with high accuracy if their increments ΔQ, $\Delta(\Delta T)$ corresponding to the increase in temperature are substituted in (10) instead of the absolute values of the quantities.

Structurally the apparatus was a stainless steel tube with an 0.15 mm wall thickness and 56 mm diameter, and having inner circular, stainless steel compression struts in the form of rings. One end of the tube was placed in a cryostat and the measuring chamber with the crystal being studied was inserted through the other end. The measuring chamber was analogous to the Liss apparatus. The tube was evacuated to 10^{-7} mm Hg, after which liquid helium was poured into the cryostat. The temperatures were measured by copper-constantan thermocouples. Thermocouples which had 0.05 mm diameter leads, were mounted on the crystal. The spacing between the thermocouples arranged on the crystal was measured to 0.001 mm accuracy by using a "katitometer." According to our computations, the error in measuring the heat conduction as a function of the temperature range was 2-5%. Presented in Fig. 1 are results on the heat conductivity of ruby, quartz, and lithium niobate, used for the computations.

The specific heat was measured on an apparatus, which is a hermetically sealed hollow vessel within which the crystal under investigation would be suspended by fine cotton threads. A manganin wire heater was wound on the crystal. The manganin wire thickness was 0.01 mm. The temperature was measured by copper-constantan thermocouples, for which the thickness of the leads was 0.05 mm. The vessel with the crystal was placed in a liquid-helium cryostat. To cool the crystal initially, the vessel was filled with gaseous helium. The vessel was evacuated to 10^{-7} mm Hg to perform the measurements. A manual potentiometer of R-306 type was used to verify the operation of the automatic potentiometer and the results of the measurements obtained.

The data on specific heat measurement were processed by means of the formula

$$C_p = \frac{Q\Delta t_{\mathrm{h}}}{m\Delta T_{\mathrm{h}}\left(1 + \frac{\Delta t_{\mathrm{h}}}{\Delta t_n}\frac{\Delta T_n}{\Delta T_{\mathrm{h}}}\right)}\,. \tag{11}$$

The method of measurement consisted in the periodic heating and subsequent cooling of the crystal. Graphs of the crystal heating and the cooling process with the heater disconnected were recorded on the tape of the automatic potentiometer. The heat losses by the crystal and the measurement errors could, respectively, be assessed from the natural cooling graphs. The errors in measurement were not more than 1%. At and near the liquid helium temperatures, the error was around 5% because of the increase in inaccuracy of the temperature measurements. The specific heat, heat conductivity, and acoustic measurements were conducted sequentially on the same specimens of material. The results of measuring the specific heat of ruby, quartz, and lithium niobate, which were used in the computations, are presented in Table 1.

TABLE 2. Dependence of the Coefficients of Hypersound Absorption α (dB/cm) on the Frequency $0.94 \cdot 10^{10}$ Hz in Ruby (Al_2O_3), Quartz (SiO_2), and Lithium Niobate ($LiNbO_3$) as a Function of Temperature (°K)

T, K	20	25	30	40	50	60	70	80	100	200
Al_2O_3	—	—	—	—	0,6	0,9	1,5	4,5	10,5	13,8
SiO_2	0,9	1,3	3,1	11,2	18,9	—	—	—	—	—
$LiNbO_3$	0,3	0,5	0,7	1,0	1,5	3,5	4,5	7,8	15,5	21

Changes in the ultrasound velocity were made in a specially fabricated electronic apparatus, described in detail in [5]. A pulse method was used to measure the velocity of ultrasound. Radio pulses of frequency up to 30 MHz and 0.3 μsec and more duration were supplied to a piezoelectric of quartz or lead zirconate—titanate (TsTS-19 or LZT-19). Specimens of different size in the form of 3 mm diameter rods or pieces of monocrystal with a several square centimeter cross-section were used. The ultrasound velocities were investigated in the 4.2-300°K temperature range. No changes in the sound velocity with temperature were hence noticed. The error in measuring the ultrasound velocities did not exceed 1%. The following values of the longitudinal and transverse ultrasound components were used in the computations: $V_l = 11.3 \cdot 10^5$ cm/sec and $V_t = 6.05 \cdot 10^5$ cm/sec for ruby; $V_l = 5.6 \cdot 10^5$ cm/sec and $V_t = 3.5 \cdot 10^5$ cm/sec for quartz; $V_l = 7.43 \cdot 10^5$ cm/sec and $V_t = 3.71 \cdot 10^5$ cm/sec for lithium niobate.

We also used the absorption coefficient we measured for hypersound at the frequency $0.94 \cdot 10^{10}$ Hz, and the absorption coefficient for hypersound at the frequency 10^9 Hz which we took from other authors [3, 6, 9], in the computations. The coaxial-resonator method [6] described earlier and the waveguide method [7] were used for the measurements. The apparatus is a combination of waveguide and resonator apparatus. The properties of the piezoeffect of the crystals themselves were used to excite hypersound in the quartz and lithium niobate, while a thin film of cadmium sulfide which possesses piezoelectric properties was deposited on the ruby. The specimen being investigated was placed with one end in the resonator and the other in the waveguide unit. The waveguide feeder and all the electronics were executed in such a manner that they permitted alternature use of two measurement methods: excitation of hypersound by using an electrical waveguide and its transmission to the resonator for recording, or to excite and record it just by a resonator (the research associated with the hypersonic investigations was performed in combination with colleagues M. A. Grigor'ev, Yu. A. Zyuryukin and V. I. Nayamov of Saratov State University). Hypersound was successfully measured from 4.2-300°K in ruby and lithium niobate. The results of measuring the value of the coefficient of hypersound absorption are presented in Table 2. The error in the absorption measurements was on the order of 10%.

DISCUSSION OF RESULTS

Successive measurements of the heat conductivity, specific heat, velocity of ultrasound, and coefficient of ultrasound absorption in the same monocrystal specimens permitted comparing the value of the heat conductivity coefficients determined phenomenologically by the Fourier method by means of (10) with the values of the heat conductivity coefficients determined on the basis of microscopic representations by the Debye formula (1) with τ_p calculated by means of (7)-(9) and V_p by means of (2). The measurements of the heat conductivity coefficients by the Fourier method are represented by solid lines in Fig. 1, and the heat conduction coefficients computed by the Debye formula are shown by points.

The good agreement between the heat conductivity data obtained by the two independent means shows that the results obtained for the phonon lifetime $\bar{\tau}_p$ from acoustic measurements are sufficiently accurate in the temperature range considered in real ruby, quartz, and lithium niobate monocrystals.

Moreover, the results obtained also show that the phonon interactions occurring in the crystals under consideration in the temperature range mentioned are actually described mainly by a process of the form $L + L \rightarrow L$. The solid lines in Fig. 2 represent the dependence of $\bar{\tau}_p$ on the temperature T computed by means of (1) in which the heat conductivity and specific heat measured by a phenomenological method enter, while the points represent the values of $\bar{\tau}_p$ computed by means of (7)-(9) on the basis of quantum acoustic theory. It is seen from Fig. 2 that starting with some temperatures, the points computed by means of (7)-(9) are somewhat higher than the values of $\bar{\tau}_p$ computed by means of (1). We assume that this is associated with the fact that phonon processes of the type $L + T_r \to T_r$ start to play a part at these temperatures, which we did not take into account in the derivation of (7)-(9). This assumption is also verified by [8] in which the deduction is made on the basis of a study of the temperature behavior of hypersound absorption coefficients in ruby, that starting with a temperature characterized by the values $(\omega\bar{\tau}_p) \sim 10$ the role of processes of $L + T_r \to T_r$ type in the absorption of hypersound is greater than that of processes of $L + L \to L$ type.

The method proposed in this paper to determine the heat conductivity of monocrystals by the classical Debye formula (1) and by using data from acoustic measurements in a broad temperature range for the phonon lifetime, therefore permits obtaining reliable results with good accuracy, and is perhaps the only one possible under specific conditions.

NOTATION

λ	is the heat conductivity;
C_V	is the specific heat;
V_p	is the mean phonon velocity;
V_l	is the velocity of the longitudinal sound wave;
V_t	is the velocity of the transverse sound wave;
$\bar{\tau}_p$	is the mean phonon lifetime;
$A(\omega_k)$	is the value of the elastic moduli of the second and third kinds;
ω	is the frequency;
$N(\omega_k)$	is the phonon density in the crystal;
ρ	is the density of the material;
T	is the temperature;
θ_D	is the value of the Debye temperature;
$\hbar$	is the Planck's constant;
Q	is the quantity of heat transmitted by the heater to the crystal;
S	is the crystal cross section;
Δx	is the spacing between points of temperature measurement;
ΔT	is the temperature drop;
Δt	is the time during which the specimen is heated to ΔT degrees;
Δt_0	is the time during which the specimen is cooled ΔT_0 degrees;
m	is the specimen mass;
L	is the phonon with longitudinal polarization;
T_r	is the phonon with transverse polarization.

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