Absorption of Sound in Insulators

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The theory of sound attenuation in structurally perfect dielectric crystals is extended and applied to recent experiments on the absorption of acoustic waves in crystalline quartz at frequencies from 10^9 cps to 2.4×10^{10} cps. The sound wave is assumed to vary the frequencies of the thermal phonons adiabatically, and the complete Boltzmann equation is used to determine the response of the thermal phonon distribution to this disturbance. The rate of energy transfer from the thermal phonons to the temperature bath is computed. In the steady state, energy is supplied by the driving sound wave to the thermal phonons at the same rate, which gives the attenuation. Relaxation times are assumed for N and U processes. Since the effect of the sound wave on a thermal phonon depends on the relative polarization and wave-number vectors of both, the phonon distribution in a small spatial region tends to relax to a new temperature T' which is determined by requiring local conservation of the total energy to first order. The present treatment leads to better understanding of the rapid decrease in attenuation with decreasing temperature in the range in which the sound-wave period becomes comparable to the average relaxation time of the thermal phonons.

1. INTRODUCTION

INTEREST in the fundamental mechanisms responsible for the damping of elastic waves (sound) in insulating crystals has recently been stimulated by the experiments of Bömmel and Dransfeld[1] and of Jacobsen,[2] in which the attenuation of such waves in quartz was studied for frequencies ranging from 10^9 to 2.4×10^{10} cps at temperatures between 4°K and 160°K. This paper is an exposition of a theory of sound absorption in insulators which provides an interpretation of these experiments. One of the results of the present work is to extend and deepen the prior discussion[1] by Bömmel and Dransfeld of their results. The interpretation given here is not always sufficiently quantitative because only very rough values of some of the parameters entering the theory are available at present, so that explicit evaluation of the complicated theoretical expressions is not always warranted. Only insulators are considered because the theory of sound absorption processes involving free electrons has already been treated in considerable detail.[3-5]

An important contribution to the microscopic theory of sound absorption was made by Landau and Rumer,[6] who presented a quantum mechanical treatment of the problem, valid when $\Omega \tau > 1$, where Ω is the circular frequency of the sound wave and τ is the mean time between collisions of a thermal phonon in the solid. They emphasized that their theory no longer applies in the range $\Omega \tau \lesssim 1$.

Akhieser[7] treated the case $\Omega \tau \ll 1$. Following the classical phenomenological analysis[8] he regarded the absorption as arising partly from heat flow and partly from viscous damping. Our work is related to and was in part suggested by his method for calculating the viscous damping contribution. We have avoided trying to separate the absorption into heat-flow and viscous damping parts because as $\Omega \tau$ approaches unity this separation is difficult to justify. The present work resembles Drude's treatment[9] of the absorption of light by electrons in its dependence on the linearized Boltzmann transport equation and heuristic assumptions as to the form of the collision integral in that equation. Like Drude's theory, it agrees fairly well with experiment for $\Omega \tau$ approaching and even greater than unity, though its derivation is rigorously valid only for $\Omega \tau < 1$.

Akhieser's calculation of the viscous damping contribution to sound absorption is based on the idea that the sound wave modulates the elastic properties and hence the thermal phonon frequencies of the medium through which it propagates. The modulated phonons are no longer in thermal equilibrium but relax towards local thermal equilibrium via phonon-phonon collisions caused by the anharmonic interaction. This relaxation is an entropy-producing process which removes energy from the sound wave driving the process and thus damps it. The time-independent Boltzmann transport equation is used to describe the process. The collision integral is written explicitly and exclusively in terms of the three-phonon transition probabilities.

The present analysis differs from that of Akhieser in that all field and time-dependent terms of the complete Boltzmann equation are considered, in keeping with the fact that the driving term has an explicit time variation. Further, the collision term is written in terms of relaxation times for normal phonon-phonon collisions (N processes) and all other processes (U processes), both of which tend to relax the distribution function to an equilibrium distribution which is characteristic of a local temperature T', and, in the case of the N processes, is shifted in phonon wave-number space away from the origin. Except for the introduction of T', this procedure is in correspondence

[1] H. E. Bömmel and K. Dransfeld, Phys. Rev. 117, 1245 (1960).
[2] E. H. Jacobsen, Phys. Rev. Letters 2, 249 (1959); article in *Quantum Electronics* (Columbia University Press, New York, 1960), p. 468.
[3] A. B. Pippard, Phil. Mag. 41, 1104 (1955).
[4] E. I. Blount, Phys. Rev. 114, 418 (1959).
[5] M. H. Cohen, M. J. Harrison, and W. A. Harrison, Phys. Rev. 117, 937 (1960).
[6] L. Landau and G. Rumer, Physik Z. Sowjetunion 11, 18 (1937).
[7] A. Akhieser, J. Phys. (U.S.S.R.) 1, 277 (1939).
[8] L. Landau and E. M. Lifshitz, *Theory of Elasticity* (Pergamon Press, London, 1959).
[9] P. Drude, Ann. Physik 1, 566 (1900).

with Callaway's treatment[10] of thermal conductivity in insulators. The temperature T' arises here because the perturbation of the distribution function by the sound wave driving the system depends on the polarization of the thermal phonons. This leads to a local relaxation of these phonons, by a process having no analog in the theory of thermal conductivity of insulators, towards a Planck distribution for which the phonons in all branches are at a common temperature T' different from the ambient temperature of the crystal. Finally, the energy transferred from the sound wave to the thermal phonons is computed differently in the present paper.

Bömmel and Dransfeld[1] based their discussion of their results on some of the ideas underlying Akhieser's calculation of the viscous damping. They emphasized the importance of the local relaxation process just described in the attenuation of the sound wave.

We turn now to a survey of the main lines of the present analysis. As suggested by Blount in his paper on ultrasonic attenuation in metals,[4] it is convenient to consider three systems: (a) the driving sound wave; (b) the dissipative system, here the assembly of thermal phonons; and (c) an external system (thermostat or heat bath) to which the thermal phonons deliver energy by means of collisions. The dissipated energy is that delivered by (a) to (b) or equivalently that delivered by (b) to (c). We study the latter process in this paper. The driving sound wave is represented as a traveling plane wave of elastic displacement of the form $\mathbf{u}_0 \cos[i(\boldsymbol{\sigma} \cdot \mathbf{r} - \Omega t)]$ where $\mathbf{u}_0$ is the amplitude and $\boldsymbol{\sigma}$ and Ω are, respectively, the wave vector and frequency of the wave. As already noted, the principal effect of this sound wave is to produce a periodic variation of the frequencies of all the thermal phonon modes of the solid with frequency Ω, which perturbs the distribution. The linearized Boltzmann transport equation is set up in Sec. 2. In Sec. 3 the rate of energy loss of the phonons to the heat bath is computed by a procedure analogous to that used by Blount.[4] In the last section (Sec. 4) the behavior of the rather complicated expressions for the attenuation Γ is examined for several important special cases.

2. THE PERTURBED THERMAL PHONON DISTRIBUTION

Our first objective is to determine the distribution function $N(\mathbf{q}; \mathbf{r},t)$ for the population of thermal phonons in a sample of unit volume. $N(\mathbf{q}; \mathbf{r},t)$ is the number of phonons of mode $\mathbf{q}$ at position $\mathbf{r}$ and time t. This approach is valid so long as $\sigma < q_{th}$, the wave number of the most abundant phonon excitation or, neglecting dispersion in the velocity of sound, so long as

$$\Omega < KT/\hbar. \tag{2.1}$$

We let the direction of sound propagation be the z axis, and, unless otherwise indicated, assume the

[10] J. Callaway, Phys. Rev. **113**, 1046 (1959).

polarization index to be included in the specification of the wave-number vectors $\mathbf{q}$ and $\boldsymbol{\sigma}$. Similarly, integration over $\mathbf{q}$ will be assumed to include summation over mode type.

The Hamiltonian for a single phonon $\mathbf{q}$ at position z at time t under the influence of the externally applied sound wave is

$$H(\mathbf{q}; z,t) = H_0(\mathbf{q}) + H_1(\mathbf{q}; z,t) = \hbar\omega(\mathbf{q}; z,t), \tag{2.2}$$

where

$$H_0 = \hbar\omega_0(\mathbf{q}), \tag{2.3}$$

$$H_1 = a(\mathbf{q}; \boldsymbol{\sigma}, \mathbf{u}_0) H_0(\mathbf{q}) \exp[i(\sigma z - \Omega t)]$$
$$= \Psi(\mathbf{q}; \boldsymbol{\sigma}, \mathbf{u}_0) \exp[i(\sigma z - \Omega t)]. \tag{2.4}$$

Thus the perturbed frequency ω is given by:

$$\omega(\mathbf{q}; z,t) = \omega_0(\mathbf{q})\{1 + a(\mathbf{q}; \boldsymbol{\sigma}, \mathbf{u}_0) \exp[i(\sigma z - \Omega t)]\}. \tag{2.5}$$

Here $\omega_0(\mathbf{q})$ is the frequency of a phonon of mode $\mathbf{q}$ in the unstrained crystal and $a(\mathbf{q}; \boldsymbol{\sigma}, \mathbf{u}_0)$ is a coefficient depending in a complicated way on $\mathbf{q}$ and $\boldsymbol{\sigma}$ and linearly on $\mathbf{u}_0$ for small amplitudes, the case considered here. The determination of the coefficient $a(\mathbf{q}; \boldsymbol{\sigma}, \mathbf{u}_0)$ is discussed in the Appendix. Throughout this paper it is understood that only the real parts of all complex expressions such as appear in (2.4) and (2.5) have physical significance. Note, too, that in general the coefficients in complex expressions [e.g., $a(\mathbf{q}; \boldsymbol{\sigma}, \mathbf{u}_0)$] are complex. We wish to distinguish three different distribution functions: the thermal equilibrium distribution at temperature T,

$$N_0(\omega_0) = [\exp(\hbar\omega_0/KT) - 1]^{-1}, \tag{2.6}$$

the local equilibrium distribution $N_0(\omega)$ corresponding to the perturbed frequencies ω, and finally the perturbed distribution $N(\mathbf{q}; z,t)$. Since in practice the attenuation is small ($\Gamma \ll \sigma$) and we are interested only in effects of lowest order in the sound amplitude, we are justified in regarding the perturbation of the phonon distribution function by the sound wave as small. Hence we can write, to first order, the perturbed distribution function in terms of the local equilibrium distribution as

$$N(\mathbf{q}; z,t) = N_0(\omega) - (N_0'/KT)\Phi(\mathbf{q}; \boldsymbol{\sigma}, \mathbf{u}_0)$$
$$\times \exp[i(\sigma z - \Omega t)], \tag{2.7}$$

where

$$N_0' \equiv dN_0(\omega_0)/d(\hbar\omega_0/KT). \tag{2.8}$$

Using Eq. (2.4) we can also relate the perturbed distribution to the thermal equilibrium distribution in first order:

$$N(\mathbf{q}; z,t) = N_0(\omega_0) + (N_0'/KT)(\Psi - \Phi)$$
$$\times \exp[i(\sigma z - \Omega t)]$$
$$\equiv N_0(\omega_0) + N_1. \tag{2.9}$$

The Boltzmann transport equation can be written

$$\left(\frac{\partial N}{\partial t}\right)_{\text{coll}} = \frac{\partial N}{\partial t} + \frac{1}{\hbar}\sum_{i=1}^{3}\left(\frac{\partial N}{\partial r_i}\frac{\partial H}{\partial q_i} - \frac{\partial N}{\partial q_i}\frac{\partial H}{\partial r_i}\right), \tag{2.10}$$

where the r_i are the position coordinates and the $\hbar q_i$ the momentum coordinates of the phonons, here considered as classical particles. That this form of Boltzmann's equation is equivalent to the more common form involving velocities and accelerations is an immediate consequence of Hamilton's equations of motion. Note that on the right-hand side of Eq. (2.10) we include the term $\partial N/\partial t$ omitted in Akhieser's work,[7] but not terms involving differentiations with respect to temperature. Omission of the term $\partial N/\partial t$ is justified, as will become clear, for $\Omega \tau \ll 1$; however, it strongly affects the results as $\Omega \tau$ approaches unity. We believe that it is incorrect to introduce the notion of temperature in connection with the Boltzmann equation, except in describing the boundary conditions or in approximating the collision integral. For the present problem (2.10) becomes

$$\left(\frac{\partial N}{\partial t}\right)_{\text{coll}} = \frac{\partial N}{\partial t} + \frac{1}{\hbar}\left(\frac{\partial N}{\partial z}\frac{\partial H}{\partial q_z} - \frac{\partial N}{\partial q_z}\frac{\partial H}{\partial z}\right); \quad (2.11)$$

upon retaining only terms of first order,

$$\left(\frac{\partial N}{\partial t}\right)_{\text{coll}} = \frac{\partial N_1}{\partial t} + \frac{1}{\hbar}\left(\frac{\partial N_1}{\partial z}\frac{\partial H_0}{\partial q_z} - \frac{\partial N_0}{\partial q_z}\frac{\partial H_1}{\partial z}\right). \quad (2.12)$$

Proceeding heuristically we next construct an approximation to the collision term. As noted, the scattering processes fall into two classes: (1) N processes, which conserve phonon wave vector, and (2) U processes which do not conserve phonon wave vector. To describe these two classes of processes we introduce the relaxation times, $\tau_N(\mathbf{q})$ associated with class (1) and $\tau_U(\mathbf{q})$ with class (2). Because they conserve wave vector, the normal processes produce relaxation towards a distribution which is shifted in phonon wave-vector space. Both kinds of processes produce relaxation towards Planck distributions which are characterized by an effective temperature

$$T'(z,t) \equiv T + \Delta T \exp[i(\sigma z - \Omega t)]. \quad (2.13)$$

$T'(z,t)$ is the temperature which would ultimately be attained in a small region around a point at position z if at time t this region were isolated from the remainder of the solid but maintained in the state of strain existing in it at that time. We can roughly specify the region around z as a sphere centered at a point having coordinate z and with radius of order $c\tau$, where c is the mean velocity of sound and τ the mean free time for the thermal phonon distribution $N(\mathbf{q};z,t)$. The distribution function towards which N processes relax may therefore be written

$$\mathfrak{N}_0(\omega; T',\lambda) \equiv \{\exp[(\hbar\omega - \lambda \cdot \mathbf{q})/KT'] - 1\}^{-1}. \quad (2.14)$$

The corresponding relaxed distribution for U processes is

$$\mathfrak{N}_0(\omega; T') \equiv \{\exp[\hbar\omega/KT'] - 1\}^{-1}. \quad (2.15)$$

The physical significance of the shifted distribution characterized by λ is discussed by Klemens[11] and Callaway.[10] Note that $\omega(\mathbf{q})$ and not $\omega_0(\mathbf{q})$ appears in the unperturbed distribution function: All collision processes tend to relax the perturbed distribution towards distributions corresponding to phonon modes of the locally strained crystal.

To first order,

$$\mathfrak{N}_0(\omega; T',\lambda) = N_0(\omega) - (N_0'/KT)[\hbar\omega_0(\Delta T/T) + \mathbf{A} \cdot \mathbf{q}]$$
$$\times \exp[i(\sigma z - \Omega t)], \quad (2.16)$$

$$\mathfrak{N}_0(\omega; T') = N_0(\omega) - (N_0'/KT)\hbar\omega_0(\Delta T/T)$$
$$\times \exp[i(\sigma z - \Omega t)], \quad (2.17)$$

where $\mathbf{A}$ is defined by

$$\lambda = \mathbf{A} \exp[i(\sigma z - \Omega t)].$$

The collision term of the Boltzmann equation is then

$$\left(\frac{\partial N}{\partial t}\right)_{\text{coll}} = -\frac{N(\mathbf{q};z,t) - \mathfrak{N}_0(\omega; T',\lambda)}{\tau_N(\mathbf{q})}$$
$$-\frac{N(\mathbf{q};z,t) - \mathfrak{N}_0(\omega; T')}{\tau_U(\mathbf{q})}$$
$$= (N_0'/KT)\{\tau_N^{-1}[\Phi - \hbar\omega_0(\Delta T/T) - \mathbf{A} \cdot \mathbf{q}]$$
$$+ \tau_U^{-1}[\Phi - \hbar\omega_0(\Delta T/T)]\}$$
$$\times \exp[i(\sigma z - \Omega t)]. \quad (2.18)$$

The parameters $\mathbf{A}$ and ΔT are determined by the following considerations: For the normal processes, wave number must be conserved, which implies:

$$\sum_j \int \mathbf{q}\left(\frac{\partial N}{\partial t}\right)_{\text{coll},N} d^3q = 0. \quad (2.19)$$

To first order, the total rate of change of the energy of the system must also vanish, implying:

$$\sum_j \int \hbar\omega_0(\mathbf{q})\left(\frac{\partial N}{\partial t}\right)_{\text{coll}} d^3q = 0. \quad (2.20)$$

In Eqs. (2.19) and (2.20) the summation over the polarization index j has been made explicit to emphasize that both integration over the Brillouin zone and summation over polarizations are involved. Non-energy-conserving processes like radiation and interactions with external surfaces are excluded throughout the present analysis. The irreversible energy changes associated with attenuation of the sound wave only occur in second order, as discussed below. The conditions (2.19) and (2.20), which refer only to a single point of coordinate z, determine ΔT in accordance with our definition of T' only when $c\tau$ is small compared to

[11] P. G. Klemens, in *Solid-State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1958), Vol. 7, p. 1.

the wavelength of sound, or $\Omega\tau < 1$. It is clear that T' approaches T as $\Omega\tau$ becomes greater than unity. This incompleteness of the conditions (2.19) and (2.20) causes no real difficulty because, as will be seen, terms in ΔT and $\mathbf{\Lambda}$ disappear from our results as $\Omega\tau$ becomes greater than unity. Further, the present analysis does not really apply in this range, since quantum mechanical effects become important. Indeed, this regime is just that to which the Landau-Rumer[6] analysis applies.

Using Eqs. (2.4), (2.9), (2.12), and (2.18) we now construct and solve the linearized Boltzmann equation. Introducing the total or combined relaxation time $\tau(\mathbf{q})$ defined by

$$[\tau(\mathbf{q})]^{-1} = [\tau_N(\mathbf{q})]^{-1} + [\tau_U(\mathbf{q})]^{-1}, \qquad (2.21)$$

we find the solution

$$\Phi = \{1 - i[\Omega - c_z(\mathbf{q})\sigma]\tau\}^{-1}\{\hbar\omega_0(\Delta T/T) \\ + (\tau/\tau_N)\mathbf{\Lambda}\cdot\mathbf{q} - i\Omega\tau\Psi\}. \qquad (2.22)$$

Upon inserting the solution for Φ into (2.18), the conditions (2.19) and (2.20) determining ΔT and $\mathbf{\Lambda}$ may be stated explicitly. We find

$$\sum_{\mathbf{q},j} \mathbf{q}(cq\tau_N)^{-1}S(\mathbf{q},j)[1 - i(\Omega - c_z\sigma)\tau]^{-1} \\ \times \{i(\Omega - c_z\sigma)\tau(\Delta T/T) + [(\tau/\tau_N) - 1 \\ + i(\Omega - c_z\sigma)\tau](\mathbf{\Lambda}\cdot\mathbf{q}/\hbar\omega_0) - i\Omega\tau a\} = 0, \quad (2.23)$$

and

$$\sum_{\mathbf{q},j} S(\mathbf{q},j)[1 - i(\Omega - c_z\sigma)\tau]^{-1} \\ \times \{i(\Omega - c_z\sigma)[(\Delta T/T) + (\tau/\tau_N)(\mathbf{\Lambda}\cdot\mathbf{q}/\hbar\omega_0)] \\ - i\Omega a\} = 0, \quad (2.24)$$

where

$$S(\mathbf{q},j) \equiv -K[\hbar\omega_0(\mathbf{q},j)/KT]^2 N_0' \qquad (2.25)$$

is the specific heat due to the mode $\mathbf{q}$, j. Equations (2.23) and (2.24) must be solved simultaneously to determine ΔT and $\mathbf{\Lambda}$. Equation (2.22) may then be used to obtain an explicit expression for Φ.

3. THE DISSIPATION

Next, following Blount's[4] approach, we calculate the rate at which energy is transferred from system (b) (thermal phonons) to system (c). We have noted already that the average rate at which energy is removed from the sound wave is equal in the steady state to the average rate Q at which energy is transferred from the phonon system to the heat bath. Q is given by

$$Q = -\sum_{\mathbf{q},j} \left\langle H\left(\frac{\partial N}{\partial t}\right)_{\text{coll}} \right\rangle, \qquad (3.1)$$

where $\langle\ \rangle$ denotes time average. In this expression the quantities H and $(\partial N/\partial T)_{\text{coll}}$ are to be understood as the real parts of the respective complex quantities previously considered. Equation (3.1) may be transformed into a more convenient form with the help of Eq. (2.11):

$$Q = -\sum_{\mathbf{q},j} \left\langle H\left[\frac{\partial N}{\partial t} + \frac{1}{\hbar}\left(\frac{\partial N}{\partial z}\frac{\partial H}{\partial q_z} - \frac{\partial N}{\partial q_z}\frac{\partial H}{\partial z}\right)\right]\right\rangle, \quad (3.2)$$

which can also be written

$$Q = \sum_{\mathbf{q},j} \left\langle \left[N\frac{\partial H}{\partial t} - \frac{\partial}{\partial t}(NH) - \frac{1}{\hbar}\frac{\partial}{\partial z}\left(NH\frac{\partial \dot{H}}{\partial q_z}\right) \\ + \frac{1}{\hbar}\frac{\partial}{\partial q_z}\left(NH\frac{\partial H}{\partial z}\right)\right]\right\rangle. \quad (3.3)$$

The second term, which is just the time derivative of the total energy, averages to zero. The fourth term vanishes when summed over the Brillouin zone.

In the present case the third term also averages to zero. However, in the presence of a uniform temperature gradient it gives rise to the entire energy dissipation. In this case,

$$\partial H/\partial z = \partial H_0/\partial z = 0. \qquad (3.4)$$

To first order, using Eq. (2.7),

$$N(\mathbf{q}; z, t) = N_0(\hbar\omega - \Phi). \qquad (3.5)$$

Then

$$\frac{\partial N}{\partial z} = N_0'\left(-\frac{\hbar\omega}{KT^2}\right)\frac{\partial T}{\partial z} - N_0\frac{1}{T}\frac{\partial T}{\partial z} + N(q; z)\frac{1}{T}\frac{\partial T}{\partial z}. \quad (3.6)$$

Only the last term gives a contribution because of the angular integration associated with $\partial H/\partial q_z = \hbar c_z(\mathbf{q})$. We thus find

$$Q' = -\frac{1}{\hbar}\sum_{\mathbf{q},j} H\frac{\partial H}{\partial q_z}\frac{\partial N}{\partial z}$$

$$= -\sum_{\mathbf{q},j} \hbar\omega_0(\mathbf{q})c_z(\mathbf{q})N(\mathbf{q},z)\frac{1}{T}\frac{\partial T}{\partial z}$$

$$= -\frac{1}{T}\mathbf{\nabla}T\cdot\mathbf{w},$$

where $\mathbf{w}$ is the thermal current. This is the well-known expression for the rate of energy dissipation by heat conduction in a uniform thermal gradient. We return now to the sound absorption problem. Here, the entire contribution to the energy dissipation comes from the first term of (3.3). For complex quantities this may be written as

$$Q = \frac{V}{16\pi^3}\sum_j \int d^3q\ \text{Re}\left\{N_1^*\frac{\partial H_1}{\partial t}\right\}. \qquad (3.7)$$

Substituting N_1 from Eqs. (2.9) and (2.22) and H_1

from Eq. (2.4), we obtain as the general expression

$$Q = -\tfrac{1}{2}T \sum_{\mathbf{q},j} S(\mathbf{q},j) \, \mathrm{Re}\left\{ \frac{i\Omega a}{1+i(\Omega-c_z\sigma)\tau} \right.$$
$$\left. \times \left[\frac{\Delta T^*}{T} + \frac{\tau}{\tau_N} \frac{\mathbf{\Lambda}\cdot\mathbf{q}}{\hbar\omega_0} + i\Omega\tau a^* \right] \right\}, \quad (3.8)$$

which will be discussed in the next section.

The attenuation Γ is given by

$$\Gamma = Q/cW, \quad (3.9)$$

where W is the energy density of the sound wave,

$$W = \rho\Omega^2 u_0^2/2. \quad (3.10)$$

ρ is the mass density.

4. DISCUSSION OF SPECIAL CASES

The expressions (3.8), (2.23), and (2.24) determine Q if the quantities $\omega_0(\mathbf{q},j)$, $\tau_N(\mathbf{q},j)$, $\tau_U(\mathbf{q},j)$, and $a(\mathbf{q};\boldsymbol{\sigma},\mathbf{u}_0)$ are known. In view of our incomplete knowledge of these functions for most materials, further consideration will be restricted to a discussion of Q in some simple limiting cases.

The angular integrations in Eq. (3.8) can be performed analytically if one assumes ω_0, τ_N, τ_U, and a to be independent of the direction of $\mathbf{q}$. They may still depend on $|\mathbf{q}|$ and on the mode type j. The angular integrals can then all be reduced to easily evaluated forms. In order, however, to obtain tractable expressions for Q, two further approximations are desirable. First, it is assumed that the solid can be represented by the Debye model, for which

$$\omega(\mathbf{q},j) = cq, \quad (4.1)$$

where c is a constant average velocity of sound. Second, τ_N, τ_U, and a are supposed independent of q. Since τ_N, τ_U, and a may be expected to be sensitive functions of $\mathbf{q}$ and j in most solids, it is clear that the second assumption causes the following results to have only qualitative significance.

The angular integrations involved in the evaluation of Eq. (3.8) are most easily expressed in terms of

$$I_{mn}(\Omega\tau) \equiv \int_{-1}^{1} \frac{\mu^m(1-\mu)^n}{1-i\Omega\tau(1-\mu)} d\mu. \quad (4.2)$$

With the preceding assumptions and definition, Eqs. (2.23), (2.24) and (3.8) become, respectively,

$$i\Omega I_{11}(\Delta T/T) + (\tau_N^{-1}-\tau^{-1})I_{20}(\Lambda/\hbar c)$$
$$+ i\Omega I_{21}(\Lambda/\hbar c) - i\Omega I_{10}a = 0, \quad (4.3)$$

$$i\Omega I_{01}(\Delta T/T) + \tau_N^{-1}I_{10}(\Lambda/\hbar c) - i\Omega I_{00}a = 0, \quad (4.4)$$

$$Q = -\tfrac{1}{4}C_v T \, \mathrm{Re}\{i\Omega a[I_{00}^*(\Delta T^*/T)$$
$$+ (\tau/\tau_N)I_{10}^*(\Lambda^*/\hbar c) + i\Omega\tau I_{00}^* a^*]\}, \quad (4.5)$$

where $\mathbf{\Lambda} = (0,0,\Lambda)$ and

$$C_v = \sum_{\mathbf{q},j} S(\mathbf{q},j) \quad (4.6)$$

is the total lattice specific heat of the solid.

It is convenient now to specialize our formulation to the case of a compressional wave, for which, as is noted in the Appendix,

$$a = -i\gamma u_0\sigma, \quad (4.7)$$

where γ is Grüneisen's constant. Then using Eqs. (3.8)–(3.10),

$$\Gamma = (C_v T\gamma^2\Omega/2\rho c^3) \, \mathrm{Im}\{ (I_{00}^*/a^*)(\Delta T^*/T)$$
$$+ (\tau I_{10}^*/\hbar c\tau_N a^*)\Lambda^* + i\Omega\tau I_{00}^* \}. \quad (4.8)$$

It is not difficult to solve Eqs. (4.3) and (4.4) for ΔT and Λ and hence determine Γ in terms of τ_N, τ_U and the other parameters. In most solids there is considerable uncertainty regarding the values of τ_N and τ_U. Their determination depends on careful experiments and analysis such as that given by Callaway[10] for germanium. We can nevertheless obtain a qualitative impression of the content of the present results by restricting consideration now to the case $(\tau_N/\tau_U)\gg 1$ and associating the remaining τ_U with the thermal conductivity relaxation time. In this limit we consider only the second of the two relaxation terms in (2.18), Λ and Eq. (4.3) being eliminated. Then Eq. (4.4) becomes

$$\Delta T/T = I_{00}a/I_{01}, \quad (4.9)$$

and

$$\Gamma = (C_v T\gamma^2\Omega/2\rho c^3) \, \mathrm{Im}\{ (I_{00}^{*2}/I_{01}) + i\Omega\tau I_{00}^* \}. \quad (4.10)$$

For $\Omega\tau \ll 1$, Eq. (4.10) becomes

$$\Gamma = C_v T\gamma^2\Omega^2\tau/3\rho c^3. \quad (4.11)$$

This result can be succinctly expressed in terms of the thermal conductivity

$$\kappa \equiv \tfrac{1}{3}C_v c^2\tau, \quad (4.12)$$

$$\Gamma = \gamma^2\Omega^2 T\kappa/\rho c^5. \quad (4.13)$$

At temperatures greater than the Debye temperature of the solid, $\kappa \sim T^{-1}$. Thus κT and hence Γ become independent of temperature, an effect that is experimentally observed in quartz.

For $\Omega\tau \gg 1$, Eq. (4.10) becomes

$$\Gamma = \pi\gamma^2\Omega C_v T/4\rho c^3. \quad (4.14)$$

It is interesting to note that this expression is independent of τ and has the same dependence on Ω and T as that arrived at by Landau and Rumer[6] in their quantum mechanical treatment of the case $\Omega\tau > 1$. From Eq. (4.10) it is seen that the term arising from ΔT is negligible in the present limit. As already pointed out in Sec. 3, the temperature shift ΔT could not be maintained with $\Omega\tau \gg 1$. If we relax for the moment the restriction $\tau_N/\tau_U \gg 1$, which eliminated the normal
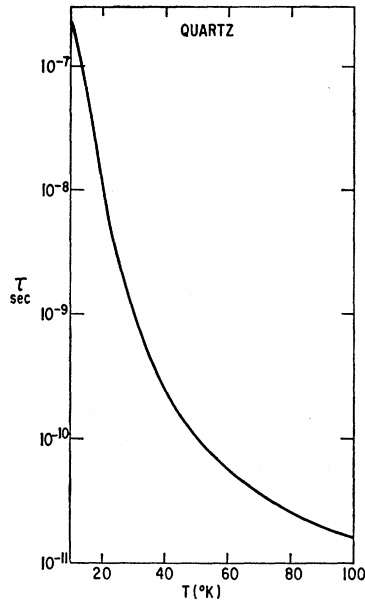
FIG. 1. The thermal conductivity relaxation time τ vs temperature. τ is computed from Eq. (4.12) using experimentally measured[12,13] values of κ and C_v.

of τ vs temperature is shown in Fig. 1. For the Debye model an average velocity of sound c can be defined in terms of the velocities c_l, c_t for longitudinal and transverse waves as follows:

$$(3/c^3) = (1/c_l^3) + (2/c_t^3). \tag{4.16}$$

The attenuation is plotted vs temperature and compared with the theoretical curves for frequencies of 1 and 3.9 kMc/sec in Figs. 2 and 3. The parameters γ are determined by matching the experimental and theoretical attenuation at 60°K in the case of the lower frequency and at 40°K for the higher frequency. Despite the grossness of the approximation involved in letting a single γ represent the coupling between the sound wave and the crystal, it is seen that the magnitude of the attenuation is represented quite well by the "refined" theory of Eq. (4.10), although the shape of the curves is represented better by the "simple" theory of Eq. (4.15).

We note that our Eq. (4.8) becomes identical with Eq. (9) of reference 1 if in addition to setting $\Delta T = \Lambda = 0$ we put $c_z(\mathbf{q}) = 0$, i.e., neglect the velocity components of the thermal phonons in the direction of the applied sound wave. This is equivalent to omitting all terms except the first on the right-hand side of Eq. (2.12). The differences between the expression in reference 1 and our result for $\Omega\tau \gg 1$ are attributable to the fact that the analysis of Bömmel and Dransfeld involves an assumption equivalent to neglect of thermal phonon velocity components in the direction of propagation of the sound wave. Our limiting expression for Γ in the $\Omega\tau \gg 1$ range is larger by a factor $\Omega\tau$, so that our considerations lead to markedly different results in this range. The difference may appear uninteresting because neither theory is really valid in this range. Neverthe-

processes, we see from Eq. (4.8) that the term depending on Λ is also unimportant in this limit. Thus Eq. (4.14) is valid in this more general case as well.

The last question concerns the form of Γ when the possibilities of the existence of finite ΔT and Λ considered here are ignored. It seems physically reasonable that relaxation should occur towards the unperturbed thermal equilibrium distribution for $\Omega\tau > 1$, when on the average a thermal phonon travels many sound wavelengths between collisions. Under these circumstances one does not expect collisions to occur often enough in any local region to alter the distribution towards which relaxation occurs. Putting $\Delta T = 0$ as well as $\Lambda = 0$ leads to the result

$$\Gamma = \frac{3\gamma^2\Omega^2 T\kappa}{\rho c^5} \frac{\tan^{-1}(2\Omega\tau)}{2\Omega\tau}, \tag{4.15}$$

which is similar to Eq. (4.10) except that the term $(I_{00}^*)^2/I_{01}$ is absent. For $\Omega\tau \ll 1$, Eq. (4.15) leads to a value of Γ three times that given by Eq. (4.13); for $\Omega\tau \gg 1$, it leads to the same value given by (4.14). Γ is considerably smaller when ΔT is considered than when ΔT is neglected (for $\Omega\tau \ll 1$) because for finite ΔT the distribution function does not have as far to relax. This results in smaller collision terms and hence smaller attenuation.

In order to check the qualitative validity of the present theory, let us compare the results given by Eqs. (4.10) and (4.15) with the experimental information available for quartz. Grüneisen's constant γ will be treated as an adjustable parameter. The thermal conductivity relaxation time will be obtained from experimental values of κ,[12] C_v,[13] and Eq. (4.12). A plot

[12] J. W. deHaas and T. Biermasz, Physica **2**, 673 (1935).
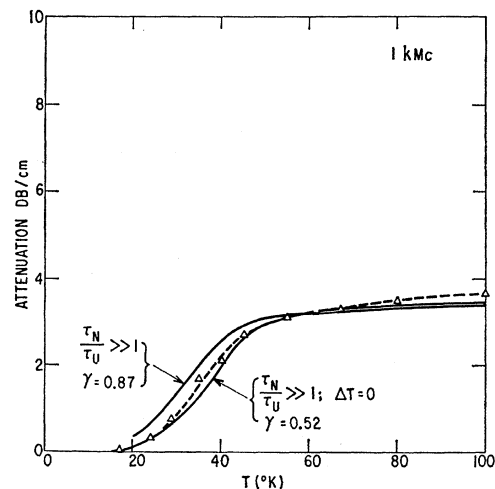[13] R. C. Lord and J. C. Morrow, J. Chem. Phys. **26**, 232 (1957).

FIG. 2. Absorption of longitudinal waves at 1.0 kMc/sec vs temperature. The dashed curve and experimentally observed points were taken from reference 1; the solid curves were obtained from the theory as described in the text.
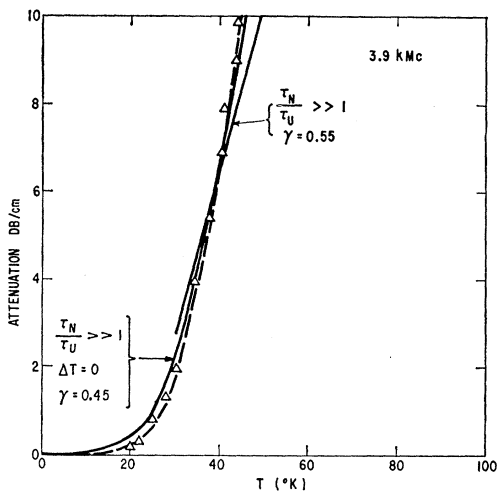
FIG. 3. Absorption of longitudinal waves at 3.9 kMc/sec vs temperature. The dashed curve and experimentally observed points were taken from reference 1; the solid curves were obtained from the theory as described in the text.

less, we remark again that except for a multiplicative constant of order unity, our result for $\Omega\tau\gg1$ is the same as that which Landau and Rumer derived from quantum mechanical arguments. For $\Omega\tau\ll1$ our result agrees with reference 1 because the terms neglected there drop out of the complete expressions in this limit.

We have seen that the present theory has the correct qualitative features and makes possible quantitative calculations for the range $\Omega\tau\ll1$, but it should be emphasized that such calculations are at least as difficult as calculations from first principles of the lattice thermal conductivity.

APPENDIX. DETERMINATION OF $a(\mathbf{q};\sigma,\mathbf{u}_0)$

We treat the calculation of $a(\mathbf{q};\sigma,\mathbf{u}_0)$ defined in Eq. (2.4), as a problem in the classical theory of elasticity. Given a displacement $\mathbf{u}(\mathbf{r})$ defined at every point $\mathbf{r}$ of the elastic solid, we seek

$$\Delta\omega(\mathbf{q};z,t)=a(\mathbf{q};\sigma,\mathbf{u}_0)\omega_0(\mathbf{q})\,\exp[i(\sigma z-\Omega t)],$$

the change in circular frequency of the phonon-packet of wave vector $\mathbf{q}$. We assume that the phonon packet is localized within a wavelength of the elastic displacement and that its frequency follows the deformation adiabatically. Both of these assumptions are valid for most of the thermal phonons if

$$\Omega<KT/\hbar. \qquad (A.1)$$

Following Akhieser,[7] we consider only the lowest order or linear dependence of $\Delta\omega(\mathbf{q};z,t)$ on the state of strain and rotation of the medium at (z,t):

$$\Delta\omega(\mathbf{q};z,t)=\omega_0(\mathbf{q})\sum_{i,k}[\gamma_{ik}(\mathbf{q})u_{ik}+\delta_{ik}(\mathbf{q})v_{ik}], \qquad (A.2)$$

with

$$u_{ik}\equiv\frac{1}{2}\left(\frac{\partial u_i}{\partial r_k}+\frac{\partial u_k}{\partial r_i}\right), \quad \text{(strain tensor)}$$

$$\qquad\qquad (A.3)$$

$$v_{ik}\equiv\frac{1}{2}\left(\frac{\partial u_i}{\partial r_k}-\frac{\partial u_k}{\partial r_i}\right), \quad \text{(rotation tensor)}$$

where r_i, $i=1$, 2, 3, are the Cartesian position coordinates and u_i are the components of the displacement. The tensor $\gamma_{ik}(\mathbf{q})$ which we call the generalized Grüneisen tensor, can be computed from the second- and third-order elastic constants of the solid and the polarization and wave vectors of mode $\mathbf{q}$ with the aid of an approximate formula given by Ziman[14] and the assumption that the effect on $\omega(\mathbf{q};r)$ of a deformation at $\mathbf{r}$ is the same as the effect on $\omega(\mathbf{q})$ of a uniform strain equal to that at $\mathbf{r}$ in a macroscopic piece of solid for which the mode $\mathbf{q}$ can be defined. The tensor $\delta_{ik}(\mathbf{q})$ relates a local rotation of the medium to the changed frequency with which a phonon of a particular wave vector and polarization would propagate in the rotated medium. In an isotropic continuum the phase velocity and frequency associated with a particular phonon are independent of the direction of propagation, and hence δ_{ik} vanishes.

We note that for a displacement described by the real part of $\mathbf{u}_0\exp[i(\sigma z-\Omega t)]$, $a(\mathbf{q};\sigma,\mathbf{u}_0)$ is a pure imaginary number. For a longitudinal wave $\mathbf{u}_0$ has only a z component, call it u_{0z}; the tensor u_{ik} has only a zz component, $u_{zz}=iu_{0z}\sigma$; and $v_{ik}=0$. Then

$$\Delta\omega=i\omega_0\gamma_{zz}u_{0z}\sigma\,\exp[i(\sigma z-\Omega t)], \qquad (A.4)$$

so that

$$a=i\gamma_{zz}u_{0z}\sigma. \qquad (A.5)$$

In this case we can identify $-\gamma_{zz}$ with the constant γ appearing in Grüneisen's theory of thermal expansion,[15] so that we have, finally,

$$a=-i\gamma u_{0z}\sigma. \qquad (A.6)$$

[14] J. M. Ziman, Electrons and Phonons (Oxford University Press, Oxford, 1960), page 154, Eq. (3.77).

[15] E. Grüneisen, Handbuch der Physik (Verlag Julius Springer, Berlin, 1926), Vol. 10, p. 21.