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Ultrasonic Attenuation in Dirty Dielectric Crystals*

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The attenuation of a sound wave by anharmonic interaction with the thermal phonons in a dielectric crystal has been calculated using the Boltzmann-equation approach of Woodruff and Ehrenreich. Contributions to the thermal-phonon collision rate due to elastic scattering of phonons by crystal defects (relaxation time τ_e) and due to intrinsic inelastic anharmonic processes (relaxation time τ_i) are considered separately. In the case of longitudinal waves the attenuation remains finite as $\tau_e \to 0$, whereas for transverse waves propagating in high-symmetry directions the attenuation tends to zero in this limit. For $\Omega\tau_i \ll 1$, an estimate is made of the ratio of the longitudinal-wave attenuation in a dirty crystal ($\Omega\tau_e \to 0$) to that in a clean crystal ($\Omega\tau_e \to \infty$). This ratio is found to be a sensitive function of the Grüneisen constants of the crystal and of the temperature.

I. INTRODUCTION

The attenuation of a high-frequency ultrasonic wave propagating in a dielectric crystal is due to an interaction between the wave and the thermal phonons.[1,2] The sound wave modulates the parameters associated with the phonons resulting in a nonequilibrium distribution. Thermal equilibrium is restored by collisions between the thermal phonons thus causing an irreversible increase of entropy and an attenuation of the wave. For $\Omega\tau \ll 1$ (Ω is the sound wave angular frequency, τ is the thermal phonon collision time) Woodruff and Ehrenreich[2] have derived the following expression for the attenuation:

$$\alpha = CT\gamma^2\Omega^2\tau/3\rho s^3, \tag{1}$$

where C is the specific heat per unit volume, γ is an average Grüneisen's constant, ρ is the density, and s is the velocity of sound. Since τ decreases with increasing temperature the condition $\Omega\tau \ll 1$ holds at high temperatures. Equation (1) has been tested experimentally[3] for a number of materials and qualitative agreement is found provided γ is treated as an adjustable parameter. There are a number of approximations involved in deriving Eq. (1), including the use of a Debye model for the phonon frequency spectrum and simplifying assumptions about the rate of collisions between phonons.

According to Eq. (1) the attenuation should be reduced if defects are added to the crystal since these will reduce the thermal phonon lifetime. If the phonon lifetime in a perfect crystal is τ_i and the phonon lifetime for elastic scattering by defects is τ_e, then the total τ is given by

$$\tau^{-1} = \tau_i^{-1} + \tau_e^{-1}. \tag{2}$$

Combining Eqs. (1) and (2) gives

$$\alpha = (CT\gamma^2\Omega^2/3\rho s^3)[\tau_i\tau_e/(\tau_i+\tau_e)]. \tag{3}$$

Hence as the number of defects increases the attenuation should decrease and in the limit $\Omega\tau_e \to 0$, $\alpha \to 0$. Experimentally a reduction in attenuation is observed but usually much smaller than would be predicted by Eq. (3). Keller[4] has made measurements of the attenuation of 640-MHz longitudinal waves in germanium-silicon alloys at 300°K. For propagation in the [100] direction the attenuation in $\mathrm{Ge}_{0.03}\,\mathrm{Si}_{0.97}$ was only 13% less than in pure silicon. The change in τ may be determined independently by measuring the thermal conductivity κ and using the approximate relation $\kappa = \frac{1}{3}Cv^2\tau$. The thermal conductivity of the alloy was found to be 8.5 times smaller than the conductivity of pure silicon and thus the τ deduced by this method was in serious disagreement with the τ necessary to explain the ultrasonic results. A similar discrepancy is found by analyzing Bömmel and Dransfeld's measurements of the attenuation in natural and neutron irradiated quartz and using the thermal conductivity as measured by Berman *et al.*[5]

In this paper we try to explain the origin of these discrepancies by considering in detail the attenuation in a crystal containing such a large number of defects that the condition $\Omega\tau_e \ll \Omega\tau_i \ll 1$ applies. We will show that, contrary to the predictions of Eq. (3), the attenuation in this limit becomes independent of $\Omega\tau_e$. The physical basis for this result is the observation that the scattering of phonons at defects is generally elastic and by itself is therefore incapable of bringing a system of phonons to complete thermal equilibrium. Thus when the condition given above applies we may consider the relaxation of the phonons after they have been disturbed by the sound wave to consist of two steps:

(i) groups of phonons having equal frequency come to mutual equilibrium in a time of the order of τ_e;

(ii) these different groups then come to equilibrium in a time τ_i, thus completing the equilibrium process.

Since the first step occurs very rapidly, the main contribution to the attenuation is from the second step and the attenuation is thus independent of τ_e.

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To calculate the attenuation we will follow the same method as Woodruff and Ehrenreich but as described above will treat the phonon collisions differently. This calculation is given in Secs. II and III. In Sec. IV we calculate the attenuation in a "clean" crystal limit $\Omega\tau_e \gg 1 \gg \Omega\tau_i$ and compare this result with the "dirty" limit to estimate the maximum reduction in attenuation that can be expected by adding defects to a crystal.

II. FORMAL EXPRESSION FOR THE ATTENUATION

We will follow the method of Woodruff and Ehrenreich[2] as adapted by Maris.[6] According to this approach the relation between the macroscopic stress tensor $\mathbf{T}$ and the strain tensor $\boldsymbol{\theta}$ is assumed to be

$$T_{\alpha\beta} = C_{\alpha\beta\gamma\delta}{}^{m}\theta_{\gamma\delta} + (\hbar/V)\sum_{\mathbf{k}j}\Omega_{\alpha\beta\gamma\delta}(\mathbf{k}j)[n(\mathbf{k}j)+\tfrac{1}{2}]\theta_{\gamma\delta} + (\hbar/V)\sum_{\mathbf{k}j}\Omega_{\alpha\beta}(\mathbf{k}j)\Delta N(\mathbf{k}j), \quad (4)$$

where $\{C_{\beta\gamma\gamma\delta}{}^{m}\}$ are second derivatives of the crystal potential energy with respect to strain and are approximately equal to the ordinary second-order elastic constants, V is the volume, $\Omega_{\alpha\beta}(\mathbf{k}j)$ and $\Omega_{\alpha\beta\gamma\delta}(\mathbf{k}j)$ are, respectively, first and second derivatives of the frequency of the normal mode $\mathbf{k}j$ with respect to strain, $n(\mathbf{k}j)$ is the thermal equilibrium number of phonons in the mode $\mathbf{k}j$, and $\Delta N(\mathbf{k}j)$ is the number of phonons in excess of the thermal equilibrium value. We adopt the convention that repeated Greek subscripts are to be summed over. The quantities involved in Eq. (4) are defined more precisely in Ref. 6. For small strains the equation of motion is

$$\frac{\partial T_{\alpha\beta}}{\partial X_\beta} = \rho\frac{\partial^2 u_\alpha}{\partial t^2}, \quad (5)$$

where $\mathbf{u}$ is the displacement at $\mathbf{X}$ and ρ is the density at the temperature of interest. We try to find a solution of Eqs. (4) and (5) of the form

$$u_\alpha = u_0 e_\alpha(\mathbf{K}J)\exp i[2\pi\mathbf{K}\cdot\mathbf{X} - \Omega(\mathbf{K}J)t], \quad (6)$$

$$\Delta N(\mathbf{k}j) = \Delta N_0(\mathbf{k}j)\exp i[2\pi\mathbf{K}\cdot\mathbf{X} - \Omega(\mathbf{K}J)t]. \quad (7)$$

Then Eq. (5) becomes

$$4\pi^2 C_{\alpha\beta\gamma\delta}{}^{m}K_\alpha K_\gamma e_\delta(\mathbf{K}J) + (4\pi^2\hbar/V)\sum_{\mathbf{k}j}\{\Omega_{\alpha\beta\gamma\delta}(\mathbf{k}j)K_\alpha K_\gamma e_\delta(\mathbf{K}J) \times[n(\mathbf{k}j)+\tfrac{1}{2}] + \Omega_{\alpha\beta}(\mathbf{k}j)(\Delta N_0(\mathbf{k}j)/2\pi i u_0)K_\alpha\} - \rho\Omega^2(\mathbf{K}J)e_\beta(\mathbf{K}J) = 0. \quad (8)$$

This equation must be solved to give $\Omega(\mathbf{K}J)$ and $\mathbf{e}(\mathbf{K}J)$. Since $\Delta N_0(\mathbf{k}j)$ will in general be complex, $\Omega(\mathbf{K}J)$ will be complex. We may relate the real and imaginary parts of $\Omega(\mathbf{K}J)$ to the velocity $s(\mathbf{K}J)$ and the attenuation α per unit length using the formulas

$$s(\mathbf{K}J) = \mathrm{Re}\,\Omega(\mathbf{K}J)/2\pi K, \quad (9)$$

$$\alpha = -\mathrm{Im}\,\Omega(\mathbf{K}J)/s(\mathbf{K}J). \quad (10)$$

The term in curly brackets in Eq. (8) corresponding to the interaction of the wave with the thermal phonons is very small compared with the other terms and may thus be treated as a perturbation. If we calculate $\mathrm{Im}\,\Omega(\mathbf{K}J)$ using standard first-order perturbation theory, we find

$$\alpha = [\pi\hbar K_\alpha e_\beta(\mathbf{K}J)/\rho V\Omega(\mathbf{K}J)s(\mathbf{K}J)] \times\sum_{\mathbf{k}j}\Omega_{\alpha\beta}(\mathbf{k}j)\,\mathrm{Re}(\Delta N_0(\mathbf{k}j)/u_0). \quad (11)$$

III. ATTENUATION IN THE DIRTY LIMIT

The problem thus reduces to calculating $\Delta N(\mathbf{k}j)$, the response of the thermal phonon system to the sound wave. To do this we follow Woodruff and Ehrenreich and only sketch their method and indicate changes in notation. The Hamiltonian of a phonon $\mathbf{k}j$ in the strain field of the sound wave is

$$H(\mathbf{k}j;\mathbf{X}t) = \hbar\omega(\mathbf{k}j;\mathbf{X}t) = \hbar[\omega(\mathbf{k}j) + \Omega_{\alpha\beta}(\mathbf{k}j)\theta_{\alpha\beta}{}^{s}(\mathbf{X}t) + \cdots], \quad (12)$$

where $\omega(\mathbf{k}j;\mathbf{X}t)$ is the frequency of a phonon $\mathbf{k}j$ at position $\mathbf{X}t$ in the presence of the sound wave and $\theta_{\alpha\beta}{}^{s}(\mathbf{X}t)$ is the strain due to the sound wave. The Boltzmann equation may be written as [Eq. (2.11) of Woodruff and Ehrenreich]:

$$\left(\frac{\partial N(\mathbf{k}j)}{\partial t}\right)_{\mathrm{coll}} = \frac{\partial N(\mathbf{k}j)}{\partial t} + \frac{1}{\hbar}\left(\frac{\partial N(\mathbf{k}j)}{\partial X_\alpha}\frac{\partial H(\mathbf{k}j;\mathbf{X}t)}{\partial k_\alpha} - \frac{\partial N(\mathbf{k}j)}{\partial k_\alpha}\frac{\partial H(\mathbf{k}j;\mathbf{X}t)}{\partial X_\alpha}\right). \quad (13)$$

$N(\mathbf{k}j) \equiv n(\mathbf{k}j) + \Delta N(\mathbf{k}j)$ is the instantaneous number of phonons of type $\mathbf{k}j$ at $\mathbf{X}$ and the left-hand side of this equation is the rate of change of $N(\mathbf{k}j)$ due to collisions. To lowest order in the amplitude of the sound wave this becomes

$$(\partial N(\mathbf{k}j)/\partial t)_{\mathrm{coll}} = \{-i\Delta N_0(\mathbf{k}j)[\Omega - 2\pi\mathbf{K}\cdot\mathbf{v}(\mathbf{k}j)] - 4\pi^2 u_0\beta\hbar n(\mathbf{k}j)[n(\mathbf{k}j)+1][\mathbf{K}\cdot\mathbf{v}(\mathbf{k}j)] \times\Omega_{\alpha\beta}(\mathbf{k}j)e_\alpha(\mathbf{K}J)K_\beta\}\exp i[2\pi\mathbf{K}\cdot\mathbf{X} - \Omega t]. \quad (14)$$

Consider now the solution of this equation when there are elastic and inelastic scattering processes. For simplicity we will not distinguish between N and U inelastic processes. The elastic scattering will relax the

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[6] H. J. Maris, Phil. Mag. **16**, 331 (1967).

distribution function towards a distribution described by a local effective "temperature" $T(\omega)$ which in general will depend on frequency. We denote this distribution by

$$N_e(\mathbf{k}j)=\{\exp[\hbar\omega(\mathbf{k}j;\mathbf{X}t)/kT(\omega(\mathbf{k}j);\mathbf{X}t)]-1\}^{-1}. \qquad (15)$$

We expand $T(\omega;\mathbf{X}t)$ as

$$T(\omega;\mathbf{X}t)=T+\Delta T(\omega)\exp i[2\pi\mathbf{K}\cdot\mathbf{X}-\Omega t]. \qquad (16)$$

If we denote the elastic scattering relaxation time by τ_e then we may approximate the rate of change of $N(\mathbf{k}j)$ due to elastic collisions by

$$(\partial N(\mathbf{k}j)/\partial t)_e=-\tau_e^{-1}[N(\mathbf{k}j)-N_e(\mathbf{k}j)]. \qquad (17)$$

The inelastic scattering processes relax the distribution function to a distribution $N_i(\mathbf{k}j)$ identical to $N_e(\mathbf{k}j)$ except that now the temperature no longer depends on frequency. We write this temperature as

$$T(\mathbf{X}t)=T+\Delta T_0\exp i[2\pi\mathbf{K}\cdot\mathbf{X}-\Omega t]. \qquad (18)$$

Then if the inelastic scattering time is τ_i,

$$(\partial N(\mathbf{k}j)/\partial t)_i=-\tau_i^{-1}[N(\mathbf{k}j)-N_i(\mathbf{k}j)]. \qquad (19)$$

If we combine Eqs. (14)–(19) we find

$$\begin{aligned}\Delta N_0(\mathbf{k}j)=\beta\hbar n(\mathbf{k}j)[n(\mathbf{k}j)+1]&\\ \times\{1+(\tau_e/\tau_i)-i[\Omega-2\pi\mathbf{K}\cdot\mathbf{v}(\mathbf{k}j)]\tau_e\}^{-1}&\\ \times\{-2\pi iu_0e_\alpha(\mathbf{K}J)K_\beta\Omega_{\alpha\beta}(\mathbf{k}j)&\\ \times[1+(\tau_e/\tau_i)+2\pi i\mathbf{K}\cdot\mathbf{v}(\mathbf{k}j)\tau_e]&\\ +[\Delta T(\omega(\mathbf{k}j))/T]\omega(\mathbf{k}j)&\\ +(\Delta T_0/T)(\tau_e/\tau_i)\omega(\mathbf{k}j)\}.& \qquad (20)\end{aligned}$$

To determine $\Delta T(\omega)$ and ΔT_0 we note that elastic scattering processes cannot change the total number of phonons having frequency in any range ω to $\omega+d\omega$. Thus

$$\sum_{\mathbf{k}j}(\partial N(\mathbf{k}j)/\partial t)_e\delta[\omega-\omega(\mathbf{k}j)]=0. \qquad (21)$$

We also have the condition that inelastic scattering processes cannot change the total energy of the phonon system. Then

$$\sum_{\mathbf{k}j}(\partial N(\mathbf{k}j)/\partial t)_i\hbar\omega(\mathbf{k}j)=0. \qquad (22)$$

If we write out these conditions explicitly using Eq. (20), we obtain

$$\begin{aligned}\tau_e^{-1}\sum_{\mathbf{k}j}\{1+(\tau_e/\tau_i)-i[\Omega-2\pi\mathbf{K}\cdot\mathbf{v}(\mathbf{k}j)]\tau_e\}^{-1}&\\ \times\delta[\omega-\omega(\mathbf{k}j)]\{2\pi\Omega\tau_eu_0e_\alpha(\mathbf{K}J)K_\beta\Omega_{\alpha\beta}(\mathbf{k}j)&\\ -[(\Delta T(\omega)-\Delta T_0)/T]\omega(\tau_e/\tau_i)&\\ +i\omega(\Delta T(\omega)/T)[\Omega-2\pi\mathbf{K}\cdot\mathbf{v}(\mathbf{k}j)]\tau_e\}=0,& \qquad (23)\end{aligned}$$

$$\begin{aligned}\tau_i^{-1}\sum_{\mathbf{k}j}\omega(\mathbf{k}j)n(\mathbf{k}j)[n(\mathbf{k}j)+1]\{1+(\tau_e/\tau_i)&\\ -i[\Omega-2\pi\mathbf{K}\cdot\mathbf{v}(\mathbf{k}j)]\tau_e\}^{-1}\{2\pi\Omega\tau_eu_0e_\alpha(\mathbf{K}J)K_\beta\Omega_{\alpha\beta}(\mathbf{k}j)&\\ +\{[\Delta T(\omega(\mathbf{k}j))-\Delta T_0]/T\}\omega(\mathbf{k}j)&\\ +i\omega(\mathbf{k}j)(\Delta T_0/T)[\Omega-2\pi\mathbf{K}\cdot\mathbf{v}(\mathbf{k}j)]\tau_e\}=0.& \qquad (24)\end{aligned}$$

Consider first the solution of these equations when $\Omega\tau_e\rightarrow 0$. Then we find that Eqs. (23) and (24) become

$$\begin{aligned}\sum_{\mathbf{k}j}\{\Omega\tau_i[2\pi u_0e_\alpha(\mathbf{K}J)K_\beta\Omega_{\alpha\beta}(\mathbf{k}j)+i\omega(\Delta T(\omega)/T)]&\\ -[(\Delta T(\omega)-\Delta T_0)/T]\omega\}\delta[\omega(\mathbf{k}j)-\omega]=0,& \qquad (25)\end{aligned}$$

$$\begin{aligned}\sum_{\mathbf{k}j}\omega^2(\mathbf{k}j)n(\mathbf{k}j)[n(\mathbf{k}j)+1]&\\ \times[\Delta T(\omega(\mathbf{k}j))-\Delta T_0]=0.& \qquad (26)\end{aligned}$$

The solution of these equations may be most simply expressed by introducing the frequency distribution $g(\omega)$ defined such that $g(\omega)d\omega$ is the fraction of normal-mode frequencies in the frequency range ω to $\omega+d\omega$. Then

$$g(\omega)=(3rN)^{-1}\sum_{\mathbf{k}j}\delta[\omega-\omega(\mathbf{k}j)],$$

where r and N are, respectively, the number of atoms per unit cell and the number of unit cells in the crystal. We also introduce a generalized Grüneisen constant $\gamma_{\alpha\beta}(\mathbf{k}j)\equiv\Omega_{\alpha\beta}(\mathbf{k}j)/\omega(\mathbf{k}j)$. The average Grüneisen constant for all modes of frequency ω is

$$\begin{aligned}\gamma_{\alpha\beta}(\omega)=\{\sum_{\mathbf{k}j}\gamma_{\alpha\beta}(\mathbf{k}j)\delta[\omega(\mathbf{k}j)-\omega]\}/&\\ \{\sum_{\mathbf{k}j}\delta[\omega(\mathbf{k}j)-\omega]\}.& \qquad (27)\end{aligned}$$

The solution of Eqs. (25) and (26) is then

$$\begin{aligned}\Delta T(\omega)/T=[(\Delta T_0/T)+2\pi u_0e_\alpha(\mathbf{K}J)K_\alpha\gamma_{\alpha\beta}(\omega)\Omega\tau_i]/&\\ (1-i\Omega\tau_i),& \qquad (28)\end{aligned}$$

$$\Delta T_0/T=2\pi iu_0e_\alpha(\mathbf{K}J)K_\beta\langle\langle\gamma_{\alpha\beta}\rangle\rangle, \qquad (29)$$

where $\langle\langle\gamma_{\alpha\beta}\rangle\rangle$ is a further average of Grüneisen constants, viz.,

$$\begin{aligned}\langle\langle\gamma_{\alpha\beta}\rangle\rangle=\int\gamma_{\alpha\beta}(\omega)\omega^2n(\omega)[n(\omega)+1]g(\omega)d\omega\Big/&\\ \int\omega^2n(\omega)[n(\omega)+1]g(\omega)d\omega,& \qquad (30)\end{aligned}$$

with $n(\omega)=[\exp(\beta h\omega)-1]^{-1}$. If we combine Eqs. (11), (20), (28), and (29) we obtain finally

$$\begin{aligned}\alpha=[2\pi^2K_\alpha K_\gamma e_\beta(\mathbf{K}J)e_\delta(\mathbf{K}J)CT/\rho s(\mathbf{K}J)]&\\ \times[\tau_i/(1+\Omega^2\tau_i^2)]&\\ \times[\langle\langle\gamma_{\alpha\beta}\gamma_{\gamma\delta}\rangle\rangle-\langle\langle\gamma_{\alpha\beta}\rangle\rangle\langle\langle\gamma_{\gamma\delta}\rangle\rangle],& \qquad (31)\end{aligned}$$

where we have introduced the specific heat C per unit volume given by

$$C=(3rN\beta\hbar^2/VT)\int_0^\infty \omega^2 n(\omega)[n(\omega)+1]g(\omega)d\omega \quad (32)$$

and

$$\langle\langle\gamma_{\alpha\beta}\gamma_{\gamma\delta}\rangle\rangle=\int \gamma_{\alpha\beta}(\omega)\gamma_{\gamma\delta}(\omega)\omega^2 n(\omega)[n(\omega)+1]g(\omega)d\omega \Big/ \int \omega^2 n(\omega)[n(\omega)+1]g(\omega)d\omega. \quad (33)$$

Equation (31) may be put in a simpler form if we introduce $\gamma_s(\mathbf{k}j)$, the effective Grüneisen constant describing the interaction of the sound wave and the mode $\mathbf{k}j$, defined as

$$\gamma_s(\mathbf{k}j)=(K_\alpha/K)e_\beta(\mathbf{k}j)\gamma_{\alpha\beta}(\mathbf{k}j). \quad (34)$$

Then we obtain finally for the attenuation in the dirty limit

$$\alpha=\frac{CT}{2\rho s^3(\mathbf{K}J)}\frac{\Omega^2\tau_i}{1+\Omega^2\tau_i^2}[\langle\langle\gamma_s^2\rangle\rangle-\langle\langle\gamma_s\rangle\rangle^2]. \quad (35)$$

$\langle\langle\gamma_s^2\rangle\rangle$ and $\langle\langle\gamma_s\rangle\rangle$ are defined analogously to $\langle\langle\gamma_{\alpha\beta}\gamma_{\gamma\delta}\rangle\rangle$ and $\langle\langle\gamma_{\alpha\beta}\rangle\rangle$.

IV. ATTENUATION IN THE CLEAN LIMIT

To be able to estimate how large an attenuation change is to be expected in going from a clean to a dirty crystal we calculate here the attenuation when $\Omega\tau_e\rightarrow\infty$. For simplicity we restrict attention to the limit $\Omega\tau_i\ll 1$ and, as in the previous section, do not distinguish between N and U processes. In this limit Eq. (23) does not apply and Eq. (24) becomes

$$\sum_{\mathbf{k}j}\omega^2(\mathbf{k}j)n(\mathbf{k}j)[n(\mathbf{k}j)+1]\{1-i[\Omega-2\pi\mathbf{K}\cdot\mathbf{v}(\mathbf{k}j)]\tau_i\}^{-1} \times\{2\pi\Omega u_0 e_\alpha(\mathbf{K}J)K_\beta\gamma_{\alpha\beta}(\mathbf{k}j) +i(\Delta T_0/T)[\Omega-2\pi\mathbf{K}\cdot\mathbf{v}(\mathbf{k}j)]\}=0.$$

To first order in $\Omega\tau_i$ this gives

$$(\Delta T_0/T)=2\pi i u_0 e_\alpha(\mathbf{K}J)K_\beta\langle\gamma_{\alpha\beta}\rangle \times[1-(i\tau_i/\Omega)\langle(2\pi\mathbf{K}\cdot\mathbf{v})^2\rangle], \quad (36)$$

where we have introduced the notation that

$$\langle f\rangle=\sum_{\mathbf{k}j}f(\mathbf{k}j)\omega^2(\mathbf{k}j)n(\mathbf{k}j)[n(\mathbf{k}j)+1]\Big/ \sum_{\mathbf{k}j}\omega^2(\mathbf{k}j)n(\mathbf{k}j)[n(\mathbf{k}j)+1]. \quad (37)$$

Then using Eqs. (11) and (20) we find to first order in $\Omega\tau_i$

$$\alpha=[CT/2\rho s^3(\mathbf{K}J)]\Omega^2\tau_i[\langle\gamma_s^2\rangle-\langle\gamma_s\rangle^2 +\langle\gamma_s\rangle^2\langle(2\pi\mathbf{K}\cdot\mathbf{v})^2\rangle/\Omega^2]. \quad (38)$$

It is straightforward to show that $\langle\gamma_s\rangle=\langle\langle\gamma_s\rangle\rangle$ but $\langle\gamma_s^2\rangle\neq\langle\langle\gamma_s^2\rangle\rangle$.

V. DISCUSSION

The result obtained in the dirty limit is clearly of the form expected from the discussion in the Introduction. In the $\Omega\tau_i\ll 1$ regime the result in the clean limit differs from the dirty limit in two ways. The averages of Grüneisen constants occurring are different and there is also an extra term. The different Grüneisen average occurs because in the clean limit the effective temperature for each individual mode relaxes by inelastic scattering towards a local average temperature. On the other hand, in the dirty case the effective temperatures of all modes with the same frequency are first averaged by elastic scattering before the inelastic scattering relaxation takes place. The last term inside the square brackets in Eq. (38) may be identified with the classical thermoelastic loss. When $\Omega\tau_i\ll 1$ the propagation of the wave is nearly adiabatic.[6] Then the *classical* attenuation due to heat conduction between the compressed and rarefied regions may be shown to be

$$\alpha=8\pi^4 T[e_\alpha(\mathbf{K}J)K_\beta C_{\alpha\beta\lambda\mu}\beta_{\lambda\mu}]^2\kappa_{\epsilon\zeta}K_\epsilon K_\zeta/ [\rho s(\mathbf{K}J)\Omega^2C^2], \quad (39)$$

where $\boldsymbol{\beta}$ and $\boldsymbol{\kappa}$ are the thermal expansion and thermal conductivity tensors, respectively. $C_{\alpha\beta\lambda\mu}$ are second-order elastic constants and we need not distinguish between isothermal and adiabatic elastic coefficients in lowest order in this equation. To demonstrate the equivalence of Eq. (39) to the "extra" term in Eq. (38) we note that by a simple extension of the results of Maradudin[7] it can be shown that

$$VC_{\alpha\beta\lambda\mu}\beta_{\lambda\mu}=-(\beta\hbar^2/T) \times\sum_{\mathbf{k}j}\gamma_{\alpha\beta}(\mathbf{k}j)\omega^2(\mathbf{k}j)n(\mathbf{k}j)[n(\mathbf{k}j)+1].$$

Using Eqs. (32) and (37) we find

$$C_{\alpha\beta\lambda\mu}\beta_{\lambda\mu}=-C\langle\gamma_{\alpha\beta}\rangle. \quad (40)$$

Equation (39) then becomes

$$\alpha=2\pi^2 T\langle\gamma_s\rangle^2\kappa_{\epsilon\zeta}K_\epsilon K_\zeta/\rho s^3(\mathbf{K}J). \quad (41)$$

To complete the equivalence we note that in the relaxation-time approximation we have used here it is straightforward to show that the thermal conductivity is

$$\kappa_{\epsilon\zeta}=\frac{\beta\hbar^2\tau_i}{VT}\sum_{\mathbf{k}j}v_\epsilon(\mathbf{k}j)v_\zeta(\mathbf{k}j)\omega^2(\mathbf{k}j)n(\mathbf{k}j)[n(\mathbf{k}j)+1] =C\tau_i\langle v_\epsilon v_\zeta\rangle. \quad (42)$$

Thus Eq. (41) becomes

$$\alpha=[CT\tau_i/2\rho s^3(\mathbf{K}J)]\langle\gamma_s\rangle^2\langle(2\pi\mathbf{K}\cdot\mathbf{v})^2\rangle, \quad (43)$$

which is equal to the extra term in Eq. (38). The reason that this term is present for the clean crystal and not

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[7] A. A. Maradudin, Phys. Status Solidi **2**, 1493 (1962).

for the dirty crystal is simply that as $\tau_e \rightarrow 0$ the thermal conductivity tends to zero and so this classical contribution to the attenuation vanishes.

How large a reduction in attenuation may be achieved by introducing defects into a crystal appears to be very dependent upon the crystal, the polarization of the sound wave, and the temperature. For cubic crystals the matrix $\boldsymbol{\gamma}(\omega)$ will be diagonal and so for shear waves propagating in high symmetry directions $\gamma_s(\omega)$ will be zero. In this case the attenuation will tend to zero as $\Omega\tau_e \rightarrow 0$. Another situation in which the attenuation will be very small in the dirty limit is at such low temperatures that only acoustic phonons near the center of the Brillouin zone are excited. In this case $\boldsymbol{\gamma}(\omega)$ will be independent of ω for all frequencies such that $n(\omega)$ is appreciable and so from Eqs. (30) and (33) we have

$$\langle\langle\gamma_{\alpha\beta}\rangle\rangle = \gamma_{\alpha\beta}(0)\,, \quad \langle\langle\gamma_{\alpha\beta}\gamma_{\gamma\delta}\rangle\rangle = \gamma_{\alpha\beta}(0)\gamma_{\gamma\delta}(0)\,,$$

where $\gamma_{\alpha\beta}(0)$ is the low-frequency limit of $\gamma_{\alpha\beta}(\omega)$. Then $\langle\langle\gamma_s\rangle\rangle^2 = \langle\langle\gamma_s^2\rangle\rangle$ and so the attenuation is again zero. Another example which is fairly simple to consider is a Debye model in which we assume that $\gamma_s(\mathbf{k}j) = \gamma_T$ for all transverse phonons and $\gamma_s(\mathbf{k}j) = \gamma_L$ for longitudinal phonons. If we denote the upper frequencies of the transverse and longitudinal branches by ω_T and ω_L, respectively, then

$$\begin{aligned} g(\omega) &= \omega^2[2\omega_T^{-3} + \omega_L^{-3}]\,, && 0 < \omega \leq \omega_T \\ &= \omega^2\omega_L^{-3}\,, && \omega_T < \omega \leq \omega_L \\ &= 0\,, && \omega_L < \omega\,. \end{aligned}$$

On this model we find that at temperatures greater than Θ_D

$$\begin{aligned} \langle\gamma_s\rangle &= \langle\langle\gamma_s\rangle\rangle = \tfrac{2}{3}\gamma_T + \tfrac{1}{3}\gamma_L\,, \\ \langle\gamma_s^2\rangle - \langle\gamma_s\rangle^2 &= (2/9)(\gamma_T - \gamma_L)^2\,, \\ \langle\langle\gamma_s^2\rangle\rangle - \langle\langle\gamma_s\rangle\rangle^2 &= (4/9)(\gamma_T - \gamma_L)^2(\omega_L^3 - \omega_T^3)/ \\ &\qquad (2\omega_L^3 + \omega_T^3)\,, \\ \langle(2\pi\mathbf{K}\cdot v)^2\rangle &= 4\pi^2K^2(\tfrac{2}{3}s_T^2 + \tfrac{1}{3}s_L^2)\,, \end{aligned}$$

where s_T and s_L are the transverse and longitudinal velocities, respectively. Then for a longitudinal sound wave and $\Omega\tau_i \ll 1$

$$\begin{aligned} \alpha_{\text{dirty}} &= (CT/9\rho s_L^3)\Omega^2\tau_i(\gamma_T - \gamma_L)^2(1 - \lambda^3)/(1 + \tfrac{1}{2}\lambda^3)\,, \\ \alpha_{\text{clean}} &= (CT/9\rho s_L^3)\Omega^2\tau_i[(\gamma_T - \gamma_L)^2 \\ &\qquad + \tfrac{1}{6}(2\gamma_T + \gamma_L)^2(1 + 2\lambda^2)]\,, \end{aligned}$$

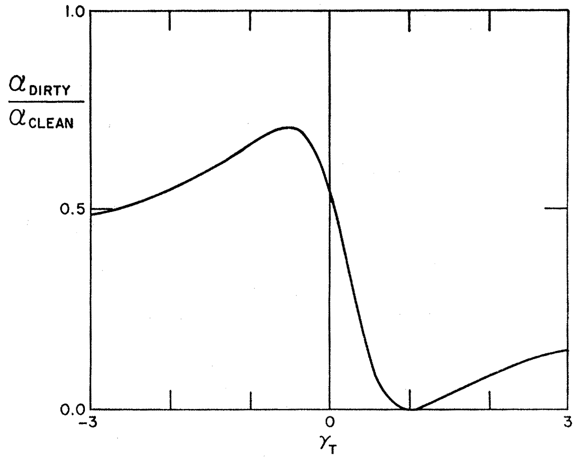

FIG. 1. Ratio of the attenuation in the dirty and clean limits as a function of the Grüneisen parameter γ_T of transverse thermal phonons. γ_L has been set equal to unity. For $\gamma_T = \gamma_L = 1$ the attenuation in the dirty limit is zero.

where $\lambda = s_T/s_L = \omega_T/\omega_L$. The ratio of these two attenuation expressions is shown as a function of γ_T in Fig. 1 for $\lambda = 0.6$ and $\gamma_L = 1$. The result is very sensitive to the ratio of γ_T to γ_L.

Finally we consider the experimental measurements of the attenuation in germanium-silicon alloys[4] and in natural and neutron-irradiated quartz[5] which were mentioned in the Introduction. It was pointed out there that these measurements were not in agreement with the simple theory [Eq. (3)] since on adding defects to a clean crystal the thermal conductivity was reduced far more than the attenuation. Quantitative comparison of these experimental results with the theory presented here is not possible because there is little information available about the Grüneisen parameters of the crystals involved and, as demonstrated in the example of the Debye model considered above, the reduction in attenuation produced by adding defects is very dependent on the details of the Grüneisen parameters. In the absence of knowledge of the Grüneisen parameters, all that can be said is that the theory does provide an explanation for the lack of correlation between these thermal conductivity and ultransonic attenuation measurements.