Acoustic Attenuation in Dielectric Solids*

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The solution of the Boltzmann equation for a phonon gas in terms of the eigenfunction spectrum of the linearized normal-process collision operator has been investigated by Guyer and Krumhansl. This treatment is extended to the case when the phonon gas is driven by an acoustic wave. An expression for the acoustic attenuation Γ is obtained in the form $\Gamma \propto \chi(\mathbf{k},\Omega)$, where $\chi(\mathbf{k},\Omega)$ is the dynamic thermal response coefficient of Griffin. $\chi(\mathbf{k},\Omega)$ depends upon the thermal conductivity $\kappa(\mathbf{k},\Omega)$, which in turn depends upon the wave vector $\mathbf{k}$ and frequency Ω. An approximation is made for $\kappa(\mathbf{k},\Omega)$ which leads to expressions for Γ in the limit $\Omega\tau_R<1$ and $\Omega\tau_R>1$ in agreement with previous investigations. In addition, in the temperature range where second sound can exist we find three frequency ranges separated by the conditions $\Omega\tau_N=1$ and $\Omega\tau_R=1$ instead of the usual two separated by $\Omega\tau_R=1$. (τ_R and τ_N are the relaxation times for non-momentum-conserving phonon scattering and normal-process scattering, respectively). The acoustic attenuation in the intermediate range, $\tau_R^{-1}\lesssim\Omega\lesssim\tau_N^{-1}$, is qualitatively different from that in the ranges $\Omega\tau_N\gtrsim 1$ and $\Omega\tau_R\lesssim 1$. Further, in this range the possibility of a resonance between first and second sound occurs. The implications of these results for acoustic attenuation experiments and the light-scattering experiment of Griffin are discussed.

I. INTRODUCTION

THE attenuation of first sound (acoustic waves) in dielectric crystals has been calculated in the limit $\Omega\tau\ll 1$ by Akhieser[1] and Ehrenreich and Woodruff,[2] and in the limit $\Omega\tau\gg 1$ by Landau and Rumer[3] and Nava *et al.*[4] The purpose of this paper is to exhibit a calculation of acoustic attenuation similar in spirit to that of Ehrenreich and Woodruff which takes advantage of the considerable simplification in treatment of the phonon Boltzmann equation introduced in the recent work of Guyer and Krumhansl.[5]

Guyer and Krumhansl have derived a set of macroscopic equations for a phonon gas with which it is possible to treat a number of phonon problems; e.g., steady-state thermal conductivity, second sound,[5] and Poiseuille flow.[6] When the phonon system is coupled to an acoustic wave (the acoustic wave is regarded as a driving force) a simple modification of the Boltzmann equation produces a modified set of macroscopic equations. These modified equations may be solved to yield the acoustic attenuation.

Recently a unified approach to interacting-phonon problems has been developed by Kwok, Martin, and Miller,[7] Kwok and Martin,[8] and Kwok.[9] In addition to reproducing most of the results of previous investigations[1–4] of acoustic attenuation, Kwok and Martin have shown the intimate relation between first sound and second sound. Both excitations correspond to poles of the single-particle Green's function and are regarded as entities of the same kind. The approach to these problems via a Boltzmann equation can in no sense reproduce the complete picture, as is possible with the procedure of Kwok and Martin. However, the Boltzmann-equation approach yields results which permit discussion of a wide range of phenomena in terms of a minimum number of phenomenological parameters, and easy comparison of the results of measurements on these phenomena.

In Sec. II we set down the Boltzmann equation that is used to describe the problem. The early stages of the calculation follow the ideas of Ehrenreich and Woodruff.[2] The Boltzmann equation is solved in Sec. III and the expression for the attenuation is obtained. The attenuation is found to depend on $\mathrm{Im}\chi(\mathbf{k},\Omega)$, where $\chi(\mathbf{k},\Omega)$ is the Griffin's dynamic thermal response coefficient,[10] and $\mathbf{k}$ and Ω are the wave number and frequency of the acoustic wave, respectively. Furthermore, $\chi(\mathbf{k},\Omega)$ depends primarily on $\kappa(\mathbf{k},\Omega)$, the $\mathbf{k}$- and Ω-dependent thermal conductivity. The major achievement of the Boltzmann-equation approach is to provide a relatively simple expression for $\kappa(\mathbf{k},\Omega)$ valid over a wide range of frequencies Ω and temperature T. In Sec. IV we examine the expression for the attenuation in the limits $\Omega\tau\ll 1$ and $\Omega\tau\gg 1$. In the limit $\Omega\tau\ll 1$ we find the usual result for the attenuation provided that $v_{\mathrm{I}}\neq v_{\mathrm{II}}$ (v_{I} and v_{II} are the velocities of first and second sound, respectively). For $v_{\mathrm{I}}\simeq v_{\mathrm{II}}$ we find that in the temperature range where second sound propagates, the acoustic wave is strongly damped. In the limit $\Omega\tau\gg 1$ we obtain an acoustic attenuation made up of a synchronous and a nonsynchronous part, in agreement with the result of Nava *et al.*[4] In Sec. V we consider the implications of our results for acoustic attenuation

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experiments and the light-scattering experiment suggested by Griffin.[10]

II. THE BOLTZMANN EQUATION

The point of view of Ehrenreich and Woodruff in treating the attenuation of first sound is the following: The attenuation of an acoustic wave in a dielectric solid is due to the transfer of energy from the wave to the phonon gas occupying the medium in which the wave is propagating. The phonon gas in turn transfers an equal amount of energy (time-averaged) to the reservoir which is in contact with the sample. The attenuation is related to the rate at which the acoustic wave loses energy to the phonons or equivalently to the rate at which the phonons lose energy to the reservoir.[11] Ehrenreich and Woodruff calculated the rate at which energy is transferred from the phonons to the reservoir. The fact that the phonons exist in a medium through which an acoustic wave is propagating is introduced into the problem by assuming that each phonon has a Hamiltonian of the form

$$H^{(\lambda)}=H_0{}^{(\lambda)}[1+\epsilon(\mathbf{x},t)]=\hbar\omega^{(\lambda)}(\mathbf{q})[1+\epsilon(\mathbf{x},t)], \quad (1)$$

where $\epsilon(\mathbf{x},t)=\epsilon_0\exp[i(\mathbf{k}\cdot\mathbf{x}-\Omega t)]$, $\mathbf{k}$ and Ω are the wave vector and frequency of the acoustic wave, respectively, λ is a polarization index, and $\epsilon(\mathbf{x},t)$ is related to the displacement of the acoustic wave u_0 by

$$\epsilon=i\gamma u_0 k_z, \quad (2)$$

where γ is the Grüneisen constant. [Equation (2) is derived in the Appendix of the paper by Ehrenreich and Woodruff.[2]] The phonons of polarization λ are then assumed to obey a single-particle Boltzmann equation with Hamiltonian given by Eq. (1), i.e.,

$$\partial N^{(\lambda)}/\partial t+[H^{(\lambda)},N^{(\lambda)}]=(\mathfrak{N}+\mathfrak{R})N^{(\lambda)}, \quad (3)$$

where $N^{(\lambda)}$ is the phonon distribution function, $[H^{(\lambda)},N^{(\lambda)}]$ is the classical Poisson bracket, and $\mathfrak{N}$ and $\mathfrak{R}$ are the operators for normal (or momentum-conserving) phonon scattering processes and non-momentum-conserving scattering processes, respectively. It is at this point that our analysis departs from that of Ehrenreich and Woodruff. These authors choose the Callaway approximation[12] to the collision operators in dealing with an equation like (3); we keep the exact linearized collision operators and deal with Eq. (3) in a manner very similar to that discussed in detail in I. Further we choose to deal with a 1-branch phonon gas for simplicity.[13] The results for a multipolarization system differ from those obtained here only in computational complexity.

When H given by Eq. (1) is introduced into Eq. (3) and N is taken to have the form

$$N=N^0(T_0)+\xi(\partial N^0/\partial\xi)\epsilon+n, \quad (4)$$

[where $\xi=\hbar\omega(q)/k_BT_0$, and $N^0(T_0)$ is the Bose-Einstein distribution at the ambient temperature of the solid, T_0], we find that n obeys the equation

$$\left(\frac{\partial}{\partial t}+\mathbf{c}\cdot\boldsymbol{\nabla}\right)n-(\mathfrak{N}+\mathfrak{R})n+\xi\frac{\partial N^0}{\partial\xi}\frac{\partial\epsilon}{\partial t}=0, \quad (5)$$

where $\mathbf{c}=\partial\omega/\partial\mathbf{q}$ is the velocity of the phonons. The second term on the right-hand side of Eq. (4) is the linearized correction to $N^0(T_0)$ which follows upon replacing $\hbar\omega(q)$ by $\hbar\omega(q)(1+\epsilon)$; the third term represents further deviation from $N_0(T_0)$ due to induced temperatures, heat currents, etc. When $\mathfrak{N}$ and $\mathfrak{R}$ are transformed into the symmetrized operators $\mathfrak{N}^*$ and $\mathfrak{R}^*$, this equation of motion becomes

$$\left(\frac{\partial}{\partial t}+\mathbf{c}\cdot\boldsymbol{\nabla}\right)n^*-(\mathfrak{N}^*+\mathfrak{R}^*)n^*+\frac{\dot{\epsilon}}{\mu}|\eta_0^*\rangle=0, \quad (6)$$

where $n^*=2(\sinh\frac{1}{2}\xi)n$. We take n^* in the form $n^*=\sum_{\nu=0}a_\nu(\mathbf{x},t)|\eta_\nu^*\rangle$, where the set of q-space functions $\{|\eta_\nu^*\rangle\}$ are the orthonormal set of eigenfunctions generated by the $\mathfrak{N}^*$ operator. A detailed treatment of Boltzmann-equation problems in this basis is given in I. One of the zero-eigenvalue eigenvectors of $\mathfrak{N}^*$ is $|\eta_0^*\rangle$ [which corresponds to a fluctuation in the phonon density, see the discussion involving Eq. (8a)],

$$|\eta_0^*\rangle=\mu\xi(2\sinh\tfrac{1}{2}\xi)^{-1},$$

so that

$$2(\sinh\tfrac{1}{2}\xi)\frac{\partial N^0}{\partial\xi}\frac{\partial\epsilon}{\partial t}\xi=-\frac{\dot{\epsilon}}{\mu}|\eta_0^*\rangle,$$

where μ is the normalization constant for $|\eta_0^*\rangle$. The combination coefficients a_ν are functions of $\mathbf{x}$ and t.

The solution of Eq. (6) leads to an expression for the attention.

In earlier work on the steady-state and the $\mathbf{k}$- and Ω- dependent thermal conductivity[5,6] (i.e., when considering phenomena in which the phonon system was not driven), the Boltzmann equation

$$(\partial/\partial t+\mathbf{c}\cdot\boldsymbol{\nabla})n^*-(\mathfrak{N}^*+\mathfrak{R}^*)n^*=0 \quad (7)$$

was employed. We see that the effect of propagating an acoustic wave through the medium occupied by the phonon system is to drive the phonon system in the $|\eta_0^*\rangle$ mode.

III. SOLUTION OF THE BOLTZMANN EQUATION

The Boltzmann equation for describing the phonon system in the undriven case, Eq. (7), leads to a set of

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[13] As long as we are concerned with the structure of the solution of the Boltzmann equation, we consider only a one-polarization system. In the computation of collision rates the true anisotropy, dispersion, and multi-polarization of the phonon spectrum could be used.

homogeneous equations for the space-time-dependent coefficients $a_\nu(\mathbf{x},t)$. We have shown that two of these, a_0 and $\mathbf{a}_1$, are simply related to quantities of physical interest.[5] The local fluctuation of the energy density is proportional to a_0, and the heat current density is proportional to $\mathbf{a}_1$. Explicitly, we found

$$\langle\epsilon\rangle-\langle\epsilon_0\rangle=(k_BT/\mu)a_0\,, \tag{8a}$$

$$Q_\alpha=\hbar k_BT(c^2/\lambda_\alpha)a_{1\alpha}\,,\quad \alpha=x,y,z \tag{8b}$$

for the energy-density fluctuation and heat current, respectively; $\langle\epsilon_0\rangle=C_VT_0/4$, μ and λ_α are the normalization constants for $|\eta_0^*\rangle$ and $|\eta_{1\alpha}^*\rangle$. Starting with the expression for the attenuation coefficient Γ from Ehrenreich and Woodruff, we can show that Γ is given by [their Eqs. (3.7), (3.9), and 3.10)]

$$\Gamma=-\tfrac{1}{3}\gamma^2\frac{C_v(T_0)T_0}{\rho v^3}\Omega\mu\,\mathrm{Im}\left(\frac{\epsilon a_0^*}{|\epsilon|^2}\right), \tag{9}$$

where $v=\Omega/k$ is the velocity of the acoustic wave. Hence, calculation of Γ depends simply upon knowledge of a_0.

We solve the equation of motion (6) in the same manner as Eq. (7) was solved in I. The solution of Eq. (6) leads to two macroscopic equations relating a_0 and the magnitude of $\mathbf{a}_1$. These are

$$\dot{a}_0+\langle 0|\mathbf{c}\cdot\boldsymbol{\nabla}|1\rangle a_1+\dot{\epsilon}/\mu=0\,, \tag{10a}$$

and

$$\langle 1|\mathbf{c}\cdot\boldsymbol{\nabla}|0\rangle a_0+\tau^{-1}a_1=0\,, \tag{10b}$$

where we use the shorthand $|0\rangle$ and $|1\rangle$ for $|\eta_0^*\rangle$ and $|\eta_1^*\rangle$, respectively, and τ^{-1} is an inner product involving a rather complicated combination of operators; it is given by

$$\tau^{-1}=\langle 1|\,(D-\mathfrak{R}^*)-(D-\mathfrak{R}^*)(D-\mathfrak{R}^*-\mathfrak{N}^*)^{-1}(D-\mathfrak{R}^*)\,|1\rangle\,, \tag{11}$$

where $D=\partial/\partial t+\mathbf{c}\cdot\boldsymbol{\nabla}$. This expression is discussed in detail in I as well as in Appendix B.

Equations (10a) and (10b) are identical with those of I for $\epsilon=0$. To calculate the $(\mathbf{k},\Omega)$-dependent thermal conductivity using (10a) and (10b), we set $\epsilon=0$ and assume that a_0 and a_1 depend upon $\mathbf{x}$ and t in the form $\exp[i(\mathbf{k}\cdot\mathbf{x}-\Omega t)]$. Then $a_1=-\langle 1|i\mathbf{k}\cdot\mathbf{c}|0\rangle a_0/\tau^{-1}$ and Eqs. (8a) and (8b) yield

$$Q_\alpha(\mathbf{k},\Omega)=-\frac{\hbar k_BT_0c^2}{\lambda_\alpha}\frac{\langle 1|i\mathbf{k}\cdot\mathbf{c}|0\rangle}{\tau^{-1}}a_0\,, \tag{12}$$

or

$$Q_\alpha(\mathbf{k},\Omega)=-\frac{\mu\hbar c^2}{\lambda_\alpha}\frac{\langle 1|i\mathbf{k}\cdot\mathbf{c}|0\rangle}{\tau^{-1}}(\langle\epsilon\rangle-\langle\epsilon_0\rangle)\,.$$

If $\langle\epsilon\rangle-\langle\epsilon_0\rangle$ is taken to be $C_v(T_0)\delta T_0\exp[i(\mathbf{k}\cdot\mathbf{x}-\Omega t)]$, Eq. (12) yields an explicit expression for the generalized thermal conductivity, $Q_\alpha(\mathbf{k},\Omega)=-ik_\alpha\kappa(\mathbf{k},\Omega)\delta T$,

$$\kappa(\mathbf{k},\Omega)=\tfrac{1}{3}C_v(T_0)c^2(1/\tau^{-1})\,. \tag{13}$$

We define $\tau(\mathbf{k},\Omega)=1/\tau^{-1}$ so that $\kappa(\mathbf{k},\Omega)$ has a simple form

$$\kappa(\mathbf{k},\Omega)=\tfrac{1}{3}C_v(T_0)c^2\tau(\mathbf{k},\Omega)\,. \tag{14}$$

The solution of Eqs. (10a) and (10b) for a_0 when $\epsilon\neq 0$ (an acoustic wave is driving the phonon system) yields

$$a_0=-\frac{\epsilon}{\mu}\left[1+\frac{\langle 0|i\mathbf{k}\cdot\mathbf{c}|1\rangle\langle 1|i\mathbf{k}\cdot\mathbf{c}|0\rangle}{i\Omega}\tau(\mathbf{k},\Omega)\right]^{-1}. \tag{15}$$

Substituting this result into Eq. (9) yields an expression for the attenuation coefficient Γ:

$$\Gamma=-\tfrac{1}{3}\gamma^2[C_v(T_0)T_0/\rho v^3]\Omega\,\mathrm{Im}\chi(\mathbf{k},\Omega)\,, \tag{16}$$

where

$$\chi(\mathbf{k},\Omega)=\left[1-\frac{k^2}{i\Omega}\frac{\kappa(\mathbf{k},\Omega)}{C_v(T_0)}\right]^{-1} \tag{17}$$

and $\kappa(\mathbf{k},\Omega)$ is given by Eq. (14). The quantity $\chi(\mathbf{k},\Omega)$ is the dynamic thermal response coefficient. Griffin has shown that the existence of a natural thermal mode (second sound) depends upon the singularities of $\chi(\mathbf{k},\Omega)$. Calculation of the attenuation of an acoustic wave involves studying the imaginary part of χ. We carry out such a calculation in the following sections. It is clear from Eq. (17) that $\kappa(\mathbf{k},\Omega)$ plays a role of primary importance; this quantity is discussed in detail in Appendix B.

We understand the result, Eq. (16), in the following way:

(a) The factor $\gamma^2C_v(T_0)T_0/\rho v^2$ is the ratio of the thermal energy density induced in the phonon system by the acoustic wave $\gamma^2C_v(T_0)T_0$ to the energy density in the acoustic wave ρv^2. The Grüneisen constant measures the coupling of the acoustic wave to the phonons.

(b) The factor $\mathrm{Im}\chi$ is proportional to the damping of the temperature wave induced by the acoustic wave.

IV. THE ACOUSTIC ATTENUATION

To calculate Γ we must evaluate $\mathrm{Im}\chi$; we write this quantity in the form

$$\mathrm{Im}\chi=\mathrm{Im}([1+\tfrac{1}{3}(c^2/v^2)i\Omega\tau(\mathbf{k}\Omega)]^{-1})\,, \tag{18a}$$

where $v=\Omega/k$ and for $\tau(\mathbf{k},\Omega)$ given by Eq. (11) we choose the following approximation:

$$\tau(\mathbf{k},\Omega)^{-1}=\langle 0|\tau_R^{-1}|0\rangle-i\Omega+[s+\langle 1|i\mathbf{k}\cdot\mathbf{c}|0\rangle]\times\langle 0|\left[i(\mathbf{k}\cdot\mathbf{c}-\Omega)+\frac{1}{\tau_N}+\frac{1}{\tau_R}\right]^{-1}|0\rangle\times[s+\langle 0|i\mathbf{k}\cdot\mathbf{c}|1\rangle]. \tag{18b}$$

In Eq. (11) D has been replaced by $i(\mathbf{k}\cdot\mathbf{c}-\Omega)$, $\mathfrak{N}^*$ and $\mathfrak{R}^*$ have been approximated by the isotropic (in q space)

scalars $-\tau_N(q)^{-1}$ and $-\tau_R(q)^{-1}$, and s is a parameter which affects $\tau(\mathbf{k},\Omega)$ only in the $\Omega\to 0$, $k\to 0$ limit, where it ensures that $\tau(0,0)$ goes to the correct combination of relaxation times to give the steady-state thermal conductivity. That is, $\tau(0,0)\to\tau_0$, where $\kappa(0,0)=\frac{1}{3}C_v(T_0)c^2\tau_0$. A method for choosing s is discussed in I and in Appendix B. As an illustration of the notation that is being used, the definition of $|\eta_0^*\rangle$ given earlier leads to

$$\langle 0|\tau_R^{-1}|0\rangle=\frac{\int_0^\infty \xi^4\frac{e^\xi}{(e^\xi-1)^2}\tau_R(\xi)^{-1}d\xi}{\int_0^\infty \xi^4\frac{e^\xi}{(e^\xi-1)^2}d\xi}=\tau_z^{-1},$$

where τ_z is the average of τ_R over the phonon spectrum corresponding to the Ziman limit.

An extensive discussion of the above approximation for $\tau(\mathbf{k},\Omega)$ is given in Appendix B.

1. $\Omega\tau\ll 1$. In this limit we consider two cases: (a) $\Omega\tau_R\ll 1$, the limit in which thermal fluctuations decay as diffusion modes, and (b) $\Omega\tau_N\ll 1$, $\Omega\tau_R\gg 1$, the limit in which thermal fluctuations decay as propagating modes.

Case (a): $\Omega\tau_R\ll 1$. In I it was shown that in this limit [see Eq. (B9)]

$$\tau(\mathbf{k},\Omega)=\tau(0,0)+O(\Omega\tau_R)+\cdots\simeq\tau_0, \tag{19}$$

where τ_0 is the relaxation time which characterizes the steady-state thermal conductivity. Further it was shown in I that examination of $\chi^{-1}=0$ to learn the character of the thermal modes leads to $\mathrm{Re}k=\mathrm{Im}k=\Omega(3/2\Omega\tau_0)^{1/2}/c$. Using Eq. (19) for $\tau(\mathbf{k},\Omega)$ in Eq. (18a) leads to $\mathrm{Im}\chi=-c^2\Omega\tau_0/3v^2$ and an acoustic attenuation given by

$$\Gamma=\tfrac{1}{3}\gamma^2(T_0\kappa_0/\rho v^5)\Omega^2. \tag{20}$$

This result is similar to Eq. (4.13) in the paper by Ehrenreich and Woodruff.[14]

Case (b): $\Omega\tau_N\ll 1$, $\Omega\tau_R\gg 1$. As with the case discussed above, this limit of the expressions for $\kappa(\mathbf{k},\Omega)$ and $\tau(\mathbf{k},\Omega)$ has been worked out in I. It was found that [see Eq. (B11)]

$$\tau(\mathbf{k},\Omega)=[i\Omega(1-i\Delta)]^{-1}, \tag{21}$$

where

$$\Delta=\tfrac{3}{5}\Omega\tau_N+(\Omega\tau_z)^{-1}. \tag{22}$$

Here τ_N stands for an average of $\tau_N(q)$ over the phonon spectrum (see Appendix B). The expression for Δ see below) can be modified to include the effect of Poiseuille flow.[6] In I it was shown that examination of $\chi^{-1}=0$ using Eq. (21) for $\tau(\mathbf{k},\Omega)$ leads to $\mathrm{Re}k=\sqrt{3}\Omega/c$, $\mathrm{Im}k=\sqrt{3}\Delta\Omega/c$, i.e., propagating thermal modes. Using Eq. (21) in Eq. (18a), we find

$$\mathrm{Im}\chi=\mathrm{Im}([1-p^2(1-i\Delta)]^{-1})\simeq\frac{-p^2\Delta}{[1-p^2]^2}, \tag{23}$$

where $p^2=c^2/3v^2\neq 1$ is the square of the ratio of the second sound velocity $v_{II}=c/\sqrt{3}$ to the first-sound velocity $v_I=v$. For $p=1$ we find $\mathrm{Im}\chi\simeq-\Delta^{-1}$. The acoustic attenuation is given by

$$\Gamma=\tfrac{1}{3}\gamma^2\frac{C_v(T_0)T_0}{\rho v^3}\Omega\frac{p^2}{[1-p^2]^2}\Delta, \quad p\neq 1, \tag{24a}$$

$$\Gamma=\tfrac{1}{3}\gamma^2\frac{C_v(T_0)T_0}{\rho v^3}\Omega\frac{1}{\Delta}, \quad p=1. \tag{24b}$$

A resonance appears in the acoustic attenuation for an acoustic wave propagating with the velocity of second sound. The possible experimental implications of this result are discussed in Sec. V. For an explanation of this resonance we note that Eq. (15) for a_0 can be written in the form

$$a_0=-(\epsilon/\mu)\chi(\mathbf{k},\Omega). \tag{25}$$

Hence, when the phonon system is driven by an acoustic wave having the velocity of second sound, a resonance in the amplitude of the induced thermal wave leads to a large transfer of energy from the acoustic wave to the phonon system.

2. $\Omega\tau\gg 1$. In order to make calculations in this limit somewhat simpler, we assume that the normal processes are the dominant phonon-scattering process. For $\tau(\mathbf{k},\Omega)$ in this limit we have [see Eq. (B12)]

$$\tau(\mathbf{k},\Omega)=\Big[-i\Omega-\langle 0|i\mathbf{k}\cdot\mathbf{c}|1\rangle\times\langle 0|\Big[i(\mathbf{k}\cdot\mathbf{c}-\Omega)+\frac{1}{\tau_N}\Big]^{-1}|0\rangle\langle 0|i\mathbf{k}\cdot\mathbf{c}|1\rangle\Big]^{-1}. \tag{26}$$

Using Eq. (26) for $\tau(\mathbf{k},\Omega)$ in Eq. (18a) we find

$$\mathrm{Im}\chi=\frac{-p^4}{(1-p^2)^2+p^4|M|^2}\mathrm{Im}M(r,\epsilon') \tag{27}$$

with

$$M(r,\epsilon')=\tfrac{1}{2}\langle 0|\int_0^\pi\frac{\sin\theta d\theta}{(r\cos\theta-1)-i\epsilon'}|0\rangle, \tag{28}$$

where $r=kc/\Omega$, $\epsilon'=[\Omega\tau_N(q)]^{-1}$. For the imaginary part of $M(r,\epsilon')$, we find

$r>1$

$$-\mathrm{Im}M(r,\epsilon')=\frac{\pi}{2r}+\langle 0|\tan^{-1}\Big(\frac{r^2}{1-r^2}\frac{1}{\Omega\tau_N}\Big)|0\rangle, \tag{29a}$$

$r<1$

$$\mathrm{Im}M(r,\epsilon')=\langle 0|\tan^{-1}\Big(\frac{r^2}{r^2-1}\frac{1}{\Omega\tau_N}\Big)|0\rangle. \tag{29b}$$

[14] In fact this equation is something of a generalization of Ehrenreich and Woodruff's (Ref. 2) Eq. (4.13); the relaxation time which they identify with the thermal conductivity relaxation time is that only at high temperature. Their Eq. (4.13) is more general than its derivation.

The term $\pi/2r$ in ImM for $r>1$ is due to a pole in the integrand of Eq. (28) which moves inside the contour. This term gives the damping of the acoustic waves due to synchronous absorption of energy by phonons which travel along with the wave, i.e., such that $r\cos\theta=1$.[15] (A physical model of this absorption mechanism has been developed by Nava *et al.*[4]) In the present limit the acoustic attenuation is given by

$$\Gamma\approx\tfrac{1}{3}\gamma^2\frac{C_v(T_0)T_0}{\rho v^3}\Omega p^4 \times\left\{\frac{\pi}{2r}\delta_{r>1}+\langle 0|\tan^{-1}\left(\frac{r^2}{|1-r^2|}\frac{1}{\Omega\tau_N}\right)|0\rangle\right\}. \quad (30)$$

The first term in curly brackets should be weighted by the ratio of the specific heat of those phonons with $r>1$ to the total specific heat; the second term in curly brackets goes as $\langle 0|(\Omega\tau_N)^{-1}|0\rangle$ for $\Omega\tau_N\to+\infty$ in agreement with the result of Nava *et al.*

V. DISCUSSION

Our primary purpose in the preceding section has been to exhibit the results of the application of Eq. (16) for the acoustic attenuation to the full range of possible experimental situations $\Omega\tau>1$ and $\Omega\tau<1$. Most of the results we have obtained are not new. However, they have been obtained from a single relatively simple expression, Eq. (16). The major statement of physical consequence in the application of this expression is the approximation Eq. (18b) for the $\mathbf{k}$- and Ω-dependent thermal conductivity $\kappa(\mathbf{k},\Omega)$ or equivalently $\tau(\mathbf{k},\Omega)$. The success of these calculations suggests that the approximation is adequate. The parameters which characterize it and thus enter the various expressions for the acoustic attenuation also characterize the wide class of thermal transport phenomena that has been discussed previously using the same approximation.[5,6] Direct comparison of experimental investigations of these various phenomena is thereby facilitated.

The major new contribution of this paper is in the treatment of acoustic attenuation in the second-sound region, i.e., the temperature and frequency region in which $\Omega\tau_N\ll1$ and $\Omega\tau_R\gg1$ are satisfied. In Fig. 1 we illustrate the qualitative Ω and T_0 dependence of the second-sound region for solid He^4 at 19.5 cm^3/mole. We consider here the possible consequences of acoustic attenuation experiments performed in this region. There are two results of interest:

(a) The condition $\Omega\tau=1$ is double-valued in the second-sound region: $\Omega\tau_N=1$, where τ_N is the time which characterizes the rate of normal-process scattering (see curve 1 on Fig. 1), and $\Omega\tau_R=1$, where τ_R is the time which characterizes non-momentum-conserving scattering (see curve 2 on Fig. 1). For frequencies satisfying the condition $1/\tau_R\lesssim\Omega\lesssim1/\tau_N$ (the shaded region in Fig. 1), the acoustic attenuation behaves differently from previous predictions. For temperatures in the second-sound region there are 3 distinct frequency ranges in which the acoustic attenuation has qualitatively different behavior.

(b) Acoustic waves which propagate with velocities near the second-sound velocity are strongly damped.

We discuss these items separately.

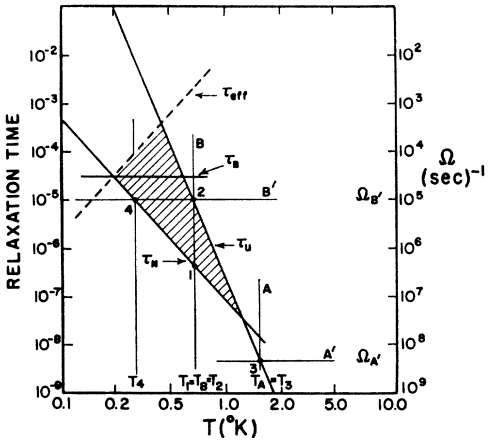

FIG. 1. Relaxation times versus temperature. The relaxation times for the various phonon-scattering mechanisms are computed from the results of the analysis of the steady-state thermal-conductivity data of Bertman *et al.* [Phys. Rev. 142, 89 (1966)]. τ_B is for a crystal of typical dimension 1 cm; $\tau_{\rm eff}$ is the effective momentum-loss relaxation time of Poiseuille flow. (See Ref. 6.)

A. Acoustic Attenuation in the Second-Sound Region

Consider an acoustic-attenuation experiment done at constant temperature over a wide range of frequencies (e.g., along the line B in Fig. 1). For very low frequencies $\Omega<10^4$ sec^{-1}, Γ is given by Eq. (20), and the attenuation is of the form

$$\frac{\Gamma}{\Omega A}=\Omega\tau_0, \quad (31a)$$

where $A=\gamma^2C_v(T_0)T_0/3\rho v^3$, and τ_0 is the time which characterizes the steady-state thermal conductivity, i.e., the momentum-non-conserving scattering processes. For frequencies between 10^4 sec^{-1} and 10^6 sec^{-1}, Γ is given by Eq. (24a):

$$\frac{\Gamma}{\Omega A}\simeq\frac{1}{\Omega\tau_0}+\tfrac{3}{5}\Omega\langle 0|\tau_N|0\rangle. \quad (31b)$$

Finally, for frequencies greater than 10^6 sec^{-1} (for which $\Omega\tau_N\simeq1$), the attenuation is given by Eq. (30),

$$\frac{\Gamma}{\Omega A}\simeq\frac{1}{\Omega}\langle 0|\tau_N^{-1}|0\rangle. \quad (31c)$$

[15] This energy-loss mechanism is the analog of Landau damping or an electron gas.

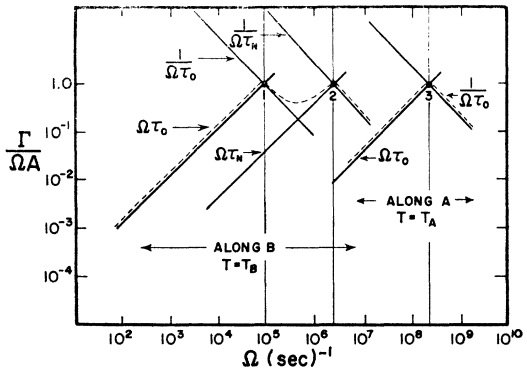

FIG. 2. Acoustic attenuation versus frequency. At temperature T_A the $\Omega\tau=1$ condition is single-valued; at T_B, $\Omega\tau=1$ is satisfied twice.

In Fig. 2 we illustrate the qualitative behavior of $\Gamma/\Omega A$ as a function of Ω for two temperatures: T_B, which passes through the second-sound region, and T_A, which is at higher temperature and should show the conventional behavior.

Consider an attenuation experiment done at constant frequency in which the temperature is swept through the second-sound region, e.g., along line B′ in Fig. 1. For temperatures above T_1 the attenuation will be given by Eq. (31a); in the intermediate temperature range, $T_4 \leqslant T \leqslant T_1$, it is given by Eq. (31b); at low temperatures it is given by Eq. (31c). In Fig. 3 we illustrate the qualitative behavior predicted by these equations for Γ over the full temperature range. The existence of the double-valued $\Omega\tau$ condition leads to the "cusp" in the behavior of Γ as a function of T and Ω. The qualitative changes in the propagation properties of the thermal wave induced by the acoustic wave which occur at $\Omega\tau_N=1$ and $\Omega\tau_0=1$ are reflected in the acoustic attenuation and lead to this "cusp."

Certainly, if there is a reasonable concentration of chemical and isotopic impurities these results are modified in an obvious way. An extensive discussion of materials in which second sound might be observed is contained in Ref. 6; the result of that discussion is that solid He^4 is by far the best material in which to look for propagating temperature wave effects.

B. Light-Scattering Experiments

Griffin[10] has recently suggested the possibility of detecting second sound in a dielectric solid using a light-scattering experiment. [The principle which permits first sound to be detected by such experiments is well known[16]]. This is the context in which we discuss the resonance in the acoustic attenuation for $v_I \simeq v_{II}$, although it is quite possible that it can be observed in a direct attenuation experiment.

[16] J. Frenkel, *Kinetic Theory of Liquids* (Clarendon Press, Oxford, England, 1946).

According to Griffin, $S(\mathbf{q},\omega)$ for a light beam scattered by a dielectric solid is given by

$$S(\mathbf{q},\omega)\approx\frac{v_I^2q^4\Gamma_I}{(\omega^2-v_I^2q^2)^2+(qv_I^2\Gamma_I)^2}+A'\frac{v_{II}^2q^4\Gamma_{II}}{(\omega^2-v_{II}^2q^2)^2+(qv_{II}^2\Gamma_{II})^2}, \quad (32)$$

where $A'=1-C_V/C_P$, and $\hbar\omega$ and $\hbar\mathbf{q}$ are the energy transfer and momentum transfer to the probe particle respectively. Eq. (32) is Griffin's Eq. (6) specialized to the region where second sound propagates. The first term in this equation is the usual result for light scattering from a medium whose density fluctuations propagate as first sound; the second term is due to light scattering from density fluctuations which accompany propagating thermal fluctuations (second sound). The propagating thermal fluctuations are the source of these density fluctuations through $\alpha=(1/V)(\partial V/\partial T)_P$; these "induced" density fluctuations obey the dispersion relation for second sound.

The amplitude of the scattering from normal density fluctuations is proportional to $\langle(\Delta V)_T^2\rangle_{\rm av}=k_BTV\beta_T$, where β_T is the isothermal compressibility of the solid. The amplitude of the thermally induced density fluctuations is proportional to $\langle(\Delta V)_P^2\rangle_{\rm av}=V^2(\partial V/\partial T)_P^2\langle(\Delta T)^2\rangle_{\rm av}$; $\langle(\Delta T)^2\rangle_{\rm av}=k_BT^2/C_V$. Hence, the ratio of the amplitude of thermally induced density fluctuations to normal density fluctuations is

$$\frac{\langle(\Delta V)_P^2\rangle_{\rm av}}{\langle(\Delta V)_T^2\rangle_{\rm av}}=\frac{\alpha^2VT}{C_V\beta_T}=\frac{C_P}{C_V}-1\simeq 1-\frac{C_V}{C_P}=A', \quad (33)$$

where we have used the thermodynamic identity $C_P-C_V=\alpha^2TV/N\beta_T$.

From Eq. (24a) the attenuation of first sound in the second-sound region (off resonance) is

$$\Gamma_I\simeq A\Omega\Delta.$$

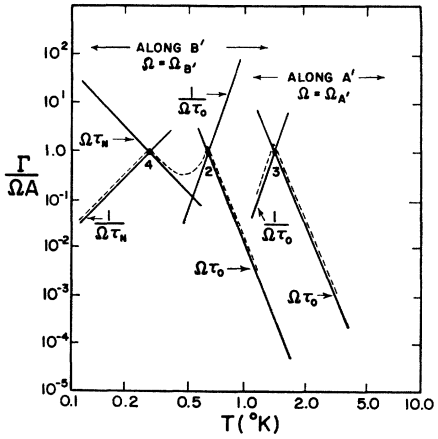

FIG. 3. Acoustic attenuation versus temperature. At $\Omega_{A'}$ the $\Omega\tau=1$ condition is single-valued; at $\Omega_{B'}$ $\Omega\tau=1$ is satisfied twice.

In the same region second sound, and hence the first sound induced by it, is damped according to [5]

$$\Gamma_{II}\simeq\Omega\Delta.$$

The ratio of Γ_{II}/Γ_I is of interest; we find

$$\frac{\Gamma_{II}}{\Gamma_I}\simeq\frac{\rho v^2}{\gamma^2 C_V(T_0)T_0}=A^{-1}=A'^{-1}$$

for a solid which obeys the Grüneisen relation.[17] The linewidths of the first- and second-sound peaks are in the inverse ratio of their amplitudes. This result is simply understood when one considers that the amplitudes of the two components of $S(\mathbf{q},\omega)$ differ by the coupling between temperature and displacement and that the dampings of first and second sound differ by the same coupling. In the second-sound region the qualitative behavior of $S(\mathbf{q},\omega)$ is adequately represented in the form

$$S(\mathbf{q},\omega)\approx\frac{v_I^2q^4A\Delta}{(\omega^2-v_I^2q^2)^2+(qv_I^2A\Delta)^2}+\frac{v_{II}^2q^4A\Delta}{(\omega^2-v_{II}^2q^2)^2+(qv_{II}^2\Delta)^2}.$$

Finally, we mention the possibility of detecting second sound by observing the velocity dependence of the damping of first sound. We have discussed this possibility in a brief note which has been already published.[18] We amend that discussion as follows: For hcp He^4 at molar volume 19.5 cm^3/mole, the average transverse phonon velocity is on the order of $\frac{1}{2}$ the average longitudinal velocity.[19] Hence, the second-sound velocity, given by the average[20]

$$v_{II}^2=\tfrac{1}{3}(C_l^{-3}+2C_t^{-3})/(C_l^{-5}+2C_t^{-5})$$

is on the order of $C_t/\sqrt{3}\simeq200$ m/sec. From the calculation of Nosanow and Werthamer,[21] we might expect the lowest velocity of a transverse acoustic wave to be on the order of 250 m/sec, i.e., it may not be possible to reach the resonance. On the other hand, off resonance [where Eq. (24a) applies] the factor $f=p^2/(1-p^2)^2$ varies from $p^2\simeq0.2$, $f\simeq0.3$ along the $(1\bar{1}1)$ direction to $p^2\simeq0.6$, $f\simeq6$ along the $(1\bar{1}0)$ direction. Hence, a variation of a factor of 20 in the width of the first-sound peak is possible as the direction of the scattering acoustic wave changes from $[1\bar{1}1]$ to $[1\bar{1}0]$. As remarked earlier[18] the full effect of the resonance can be a factor as great as 100 in the width of the first-sound peak. It is clear from the above discussion that in order to take advantage of the possibility of observing the full resonance we must have materials with considerably more longitudinal specific heat than He^4 and at least as great an anisotropy.

APPENDIX A

In this Appendix we review briefly the results reported in I for the solution of the Boltzmann equation, Eq. (3), using the set of eigenfunctions generated by the symmetrized normal-process collision operator.

The Boltzmann equation

$$\partial n^*/\partial t+\mathbf{c}\cdot\nabla n^*=(\mathfrak{N}^*+\mathfrak{R}^*)n^* \tag{A1}$$

is solved by writing $n^*=\sum_\nu a_\nu(\mathbf{x},t)|\eta_\nu^*\rangle$, where the set of q-space eigenfunctions $\{\eta_\nu^*\}$ obey the equations

$$\mathfrak{N}^*|\eta_\nu^*\rangle=\lambda_\nu|\eta_\nu^*\rangle,\quad\langle\eta_\mu^*|\eta_\nu^*\rangle=\delta_{\mu\nu}.$$

Since the normal processes conserve energy and momentum, there are 4 eigenfunctions of $\mathfrak{N}^*$ which have zero eigenvalue. These are

$$|\eta_0^*\rangle=\mu\xi(2\sinh\tfrac{1}{2}\xi)^{-1}, \tag{A2}$$

$$|\eta_{1\alpha}^*\rangle=(\lambda_\alpha q_\alpha/k_BT)(2\sinh\tfrac{1}{2}\xi)^{-1}. \tag{A2}$$

As we have reported in the text, the energy density fluctuation and heat-current components are directly proportional to a_0 and $a_{1\alpha}$ respectively, i.e., to

$$\langle\eta_0^*|\eta^*\rangle=a_0\quad\text{and}\quad\langle\eta_{1\alpha}^*|n^*\rangle=a_{1\alpha}.$$

The solution of Eq. (A1) takes the form of two macroscopic equations; these are

$$\dot{a}_0+\langle0|\mathbf{c}\cdot\nabla|1\rangle a_1=0, \tag{A3a}$$

and

$$\langle1|\mathbf{c}\cdot\nabla|0\rangle a_0+\tau^{-1}a_1=0. \tag{A3b}$$

The first of these equations is the energy conservation equation; the second is a generalized thermal conductivity relation [see Eqs. (12) and (13) of the text]. The nontrivial physical content of these equations is in

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[17] Solid He^4 obeys the Grüneisen relation extremely well (J. Jarvis, private communication).

[18] R. A. Guyer, Phys. Letters **19**, 261 (1965).

[19] J. H. Vignos and H. A. Fairbank, in *Proceedings of the Eighth International Conference on Low Temperature Physics, London, 1962*, edited by R. O. Davis (Butterworth Scientific Publications, Ltd., London, 1962). F. P. Lipschultz and D. M. Lee, Phys. Rev. Letters **14**, 1017 (1965). The procedure for forming the sample in both of these experiments was such that it is highly probable that they are done on polycrystalline samples.

[20] The major contribution of a multipolarization system in modifying the results obtained is in the expression for the second-sound velocity. See, for example, J. A. Sussmann and A. Thellung, Proc. Phys. Soc. **81**, 1122 (1963).

[21] L. H. Nosanow and N. R. Werthamer, Phys. Rev. Letters **15**, 618 (1965).

the specification of τ^{-1}. In detail τ^{-1} is given by

$$\tau^{-1}=\langle 1|D-R^*|1\rangle-\sum_{\nu=2;\mu=2}\langle 1|D-R^*|\nu\rangle \times\langle\nu|(D-\mathfrak{R}^*-\mathfrak{N}^*)^{-1}|\mu\rangle\langle\mu|D-\mathfrak{R}^*|1\rangle, \quad \text{(A4)}$$

where $D=\partial/\partial t+\mathbf{c}\cdot\nabla$. The sums on ν and μ start at $\mu=2$, $\nu=2$, i.e., none of the null-space components of $\mathfrak{N}^*$ are involved. An approximation to τ^{-1} which we believe preserves the essential physical content of Eq. (A4) is discussed in Appendix B.

APPENDIX B

The explicit equation for $\tau(\mathbf{k},\Omega)$ is

$$\tau(\mathbf{k},\Omega)^{-1}=\langle\eta_{1\alpha}^*|-i\Omega+\tau_R^{-1}|\eta_{1\alpha}^*\rangle - \sum_{\mu=2;\nu=2}\langle\eta_{1\alpha}^*|i\mathbf{k}\cdot\mathbf{c}+\tau_R^{-1}|\eta_\nu^*\rangle \times\langle\eta_\nu^*|\left[i(\mathbf{k}\cdot\mathbf{c}-\Omega)+\frac{1}{\tau_R}+\frac{1}{\tau_N}\right]^{-1}|\eta_\mu^*\rangle \times\langle\eta_\mu^*|i\mathbf{k}\cdot\mathbf{c}+\tau_R^{-1}|\eta_{1\alpha}^*\rangle, \quad \text{(B1)}$$

where D has been replaced by $i(\mathbf{k}\cdot\mathbf{c}-\Omega)$, and the scattering operators $\mathfrak{R}^*$ and $\mathfrak{N}^*$ have been replaced by the isotropic scalar functions of $\mathbf{q}$, $-\tau_R(\mathbf{q})$ and $-\tau_N(\mathbf{q})$, respectively.

In the limit as $|\mathbf{k}|\to\Omega\to 0$, Eq. (B1) takes the form

$$\tau(0,0)^{-1}=\langle\eta_{1\alpha}^*|\tau_R^{-1}|\eta_{1\alpha}^*\rangle - \sum_{\mu=2;\nu=2}\langle\eta_{1\alpha}^*|\tau_R^{-1}|\eta_\nu^*\rangle\langle\eta_\nu^*|\left[\frac{1}{\tau_R}+\frac{1}{\tau_N}\right]^{-1}|\eta_\mu^*\rangle \times\langle\eta_\mu^*|\tau_R^{-1}|\eta_{1\alpha}^*\rangle. \quad \text{(B2)}$$

An acceptable approximation to this result was found in I; there a single vector replaced the sum over ν and μ, i.e.,

$$\sum_{\nu=2;\mu=2}\langle\eta_{1\alpha}^*|\tau_R^{-1}|\nu\rangle\langle\nu|\left[\frac{1}{\tau_R}+\frac{1}{\tau_N}\right]^{-1}|\mu\rangle\langle\mu|\tau_R^{-1}|\eta_{1\alpha}^*\rangle\to \langle\eta_{1\alpha}^*|\tau_R^{-1}|2\rangle\langle 2|\left[\frac{1}{\tau_R}+\frac{1}{\tau_N}\right]^{-1}|2\rangle\langle 2|\tau_R^{-1}|\eta_{1\alpha}^*\rangle = s^2\langle\eta_0^*|\left[\frac{1}{\tau_R}+\frac{1}{\tau_N}\right]^{-1}|\eta_0^*\rangle,$$

where we have defined $s=\langle\eta_{1\alpha}^*|\tau_R^{-1}|2\rangle$ a parameter to be determined by physical considerations. The matrix element $\langle 2|\tau_c|2\rangle$ has been approximated by $\langle 0|\tau_c|0\rangle$; $\tau_c^{-1}=\tau_R^{-1}+\tau_N^{-1}$. Equation (B2) takes the form

$$\tau(0,0)^{-1}=\langle\eta_{1\alpha}^*|\tau_R^{-1}|\eta_{1\alpha}^*\rangle-s^2\langle\eta_0^*|\tau_c|\eta_0^*\rangle. \quad \text{(B3)}$$

Using the definition of $|\eta_0^*\rangle$ and $|\eta_{1\alpha}^*\rangle$ we have

$$\langle\eta_{1\alpha}^*|\tau_R^{-1}|\eta_{1\alpha}^*\rangle=\frac{\int x^4\frac{e^x}{(e^x-1)^2}\tau_R(x)^{-1}dx}{\int x^4\frac{e^x}{(e^x-1)^2}dx}=\tau_z^{-1} =\langle\eta_0^*|\tau_R^{-1}|\eta_0^*\rangle=\langle 0|\tau_R^{-1}|0\rangle, \quad \text{(B4)}$$

i.e., the resistive relaxation time computed in the Ziman limit, and

$$\langle\eta_0^*|\tau_c|\eta_0^*\rangle=\int x^4\frac{e^x}{(e^x-1)^2}\tau_c(x)dx\Big/\int x^4\frac{e^x}{(e^x-1)^2}dx. \quad \text{(B5)}$$

We know that in the Ziman limit $\tau_N\to 0$ and $\tau(0,0)\to\tau_z$. This is in agreement with Eq. (B3). In the opposite limit (the Debye limit) $\tau_R\to 0$, we know from physical considerations that $\tau(0,0)\to\tau_D$, where

$$\tau_D=\int x^4\frac{e^x}{(e^x-1)^2}\tau_R(x)dx\Big/\int x^4\frac{e^x}{(e^x-1)^2}dx. \quad \text{(B6)}$$

We choose the parameter s in such a way as to ensure that $\tau_R\to 0$ Eq. (B3) leads to $\tau(0,0)\to\tau_D$. This requirement determines s. We find

$$s^2=\frac{[\langle 0|\tau_R|0\rangle^{-1}-\langle 0|\tau_R^{-1}|0\rangle]}{\langle 0|\tau_c|0\rangle}, \quad \text{(B7)}$$

so that $\tau(0,0)$ becomes

$$\tau(0,0)=|[\langle 0|\tau_R^{-1}|0\rangle(1-s_c)+\langle 0|\tau_R|0\rangle^{-1}s_c]^{-1}, \quad \text{(B8)}$$

where $s_c=\langle 0|\tau_c|0\rangle/\langle 0|\tau_R|0\rangle$. This result is similar to but not the same as that in I. We suggest that the present experimental situation with respect to steady-state thermal conductivity is not sufficiently clean to discriminate between a number of qualitatively similar expressions for $\tau(0,0)$.[6]

The time $\tau(0,0)$ given by Eq. (B8) is the time which characterizes the steady-state thermal conductivity, i.e., $\kappa(0,0)=\frac{1}{3}C_V(T_0)c^2\tau(0,0)=\frac{1}{3}C_V(T_0)c^2\tau_0$.

We are interested in $\tau(\mathbf{k},\Omega)$ away from the $\Omega\to 0$ limit. For example consider the limit $\Omega\tau_R\ll 1$. We find that in this limit $\tau(\mathbf{k},\Omega)$ is different from $\tau(0,0)$ only in order $\Omega\tau_R$. Hence, (for $\Omega\tau_R\ll 1$)

$$\tau(\mathbf{k},\Omega)=\tau(0,0)+O(\Omega\tau_R)+\cdots. \quad \text{(B9)}$$

On the other hand, if $\Omega\tau_R\gg 1$ but $\Omega\tau_N\ll 1$, a very different circumstance exists. In the spirit of the approximation made above, Eq. (B1) becomes

$$\tau(\mathbf{k},\Omega)^{-1}=-i\Omega+\tau_z-\langle 1|i\mathbf{k}\cdot\mathbf{c}|0\rangle\langle 0|\tau_N|0\rangle\langle 0|i\mathbf{k}\cdot\mathbf{c}|1\rangle, \quad \text{(B10)}$$

or upon computing the matrix elements

$$\tau(\mathbf{k},\Omega)^{-1}=-i\Omega(1-i\Delta), \quad \text{(B11)}$$

where

$$\Delta=\tfrac{2}{5}\Omega\tau_N+(\Omega\tau_z)^{-1}.$$

In the limit $\Omega\tau_N\gg1$, $\tau_R\gg\tau_N$, the above approximation leads to

$$\tau(\mathbf{k},\Omega)=-i\Omega-\langle1|i\mathbf{k}\cdot\mathbf{c}|0\rangle \times\langle0|\left[i(\mathbf{k}\cdot\mathbf{c}-\Omega)+\frac{1}{\tau_N}\right]^{-1}|0\rangle\langle0|i\mathbf{k}\cdot\mathbf{c}|1\rangle. \quad \text{(B12)}$$

Equations (B8), (B9), (B11), and (B12) are the expressions for $\tau(\mathbf{k},\Omega)$ which are used in the text. They can be conveniently summarized in a single approximate expression for $\tau(\mathbf{k},\Omega)$. This expression is

$$\tau(\mathbf{k},\Omega)^{-1}=-i\Omega+\tau_z+[s+\langle1|i\mathbf{k}\cdot\mathbf{c}|0\rangle]^2 \times\langle0|\left[i(\mathbf{k}\cdot\mathbf{c}-\Omega)+\frac{1}{\tau_N}+\frac{1}{\tau_R}\right]^{-1}|0\rangle, \quad \text{(B13)}$$

where $s=\langle0|\tau_N|0\rangle/\langle0|\tau_R|0\rangle$. It is this approximation to $\tau(\mathbf{k},\Omega)$ or equivalently $\kappa(\mathbf{k},\Omega)$ which leads to a correct expression for steady-state thermal conductivity, the existence of second sound, and the wide range of acoustic damping phenomena discussed in the text.

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F-Aggregate Centers in NaCl: Vibronic Structure and Symmetry Properties*

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Most color-center bands associated with *F*-aggregate centers in alkali halides show relatively strong 0-phonon lines and vibronic structure when observed at low temperature. The vibronic peak energies can in almost all cases be closely correlated on an empirical basis with zone-boundary critical-point energies of normal lattice phonon modes. However, without detailed knowledge of the defect geometries and electronic states involved, it is impossible to predict what the electron-phonon coupling should be. We present the observed vibronic spectrum for all the centers in NaCl which we have found to show fine structure, and discuss the apparent electron-phonon coupling seen. As a first step toward developing models for these centers, we also present results of stress splitting measurements on the 0-phonon lines in each case. The 0-phonon lines we have studied, their band assignments, and their observed symmetry as interpreted in terms of removal of orientational degeneracy, are as follows: 6329 Å, R_2, trigonal; 8375 Å, N_1, monoclinic I; 8681 Å, N_2, monoclinic I; 1.219 μ, ?, monoclinic I; 1.306 μ, M^+, monoclinic I; 1.404 μ, impurity, rhombic I; 1.549 μ, ?, rhombic I. In several cases the relative intensities of the line components can be analyzed to give the orientation of the dipole moment involved in the 0-phonon transition. Where it is possible to propose models by combining our stress measurements with the results of other experiments, we do so, with discussion as to the probable merit of each model.

I. INTRODUCTION

THE present state of knowledge of the group of color centers in the alkali halides known as the *F*-aggregate centers is quite meager. With the exception of the simplest of them, the *R* and *M* centers, this lack of knowledge extends even to almost complete uncertainty concerning the number and geometrical arrangement of the *F* centers which are assumed to coalesce to form them.[1] In addition, almost nothing is known about the details of their interaction with the surrounding lattice and each other. However, a particularly attractive approach to obtaining much of this information is available. This is due to the fact that many of the centers in this group are coupled to the lattice with an intermediate strength such that a relatively strong 0-phonon line and more or less detailed vibrational side-band structure is visible at low temperature. The presence of this fine structure makes a number of measurements feasible which are not possible with the usual broad band alone.

It has been observed in all cases where fine structure is present on the *F*-aggregate bands of the alkali halides that the numerical values of almost all of the vibrational peak energies, measured with respect to the 0-phonon line positions, correspond very closely to normal lattice phonon mode energies near Brillouin zone boundary critical points in high-symmetry directions.[2] This strongly suggests that the electron-phonon interaction at these centers is primarily with normal lattice modes rather than local modes, and that the coupling parameters are strongly peaked at the critical

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[1] For a recent survey of this field see, e.g., W. Dale Compton and Herbert Rabin, *Solid State Physics* (Academic Press Inc., New York, 1964), Vol. 16, p. 121.
[2] C. B. Pierce, Phys. Rev. 135, A83 (1964).