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## Heat-Pulse Propagation in Dielectric Solids\*

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The usual phonon Boltzmann equation is solved by using two mean relaxation times,  $\tau_N$  for normal and  $\tau_R$  for resistive processes. For a Debye solid with three polarizations, an explicit expression for the Fourier transform of the local temperature in a heat-pulse experiment is calculated. It describes hydrodynamic phenomena for  $\Omega\tau \ll 1$ , such as second sound and diffusive heat conduction, and heat transport by ballistic phonons for  $\Omega\tau \gg 1$ . In the intermediate regime,  $\Omega\tau \approx 1$ , we find the following results: a second-sound wave with wave vector  $\vec{Q}$  can only propagate if  $Q\tau_N$  and  $Q\tau_R$  are smaller than certain critical values,  $(Q\tau_N)_c$  and  $(Q\tau_R)_c$ , i.e., for  $T \geq T_c$ , assuming the usual monotonic  $T$  dependence of  $\tau_N$  and  $\tau_R$ . The velocity  $C_2$  of second sound strongly depends on these relaxation times. Its maximum value, occurring at  $T = T_c$ , is the larger the smaller the ratio  $(\tau_N)_c/(\tau_R)_c$ . Then  $C_2$  decreases with rising  $T$  and finally goes to zero for  $\Omega\tau_R \lesssim 1$ .

### I. INTRODUCTION

In a solid the condition for second sound to be observable is the well-known frequency window

$$\bar{\Omega}\bar{\tau}_N \ll 1 \ll \bar{\Omega}\bar{\tau}_R, \quad (1.1)$$

where  $\bar{\Omega}$  is the frequency of the second-sound oscillation, and  $\bar{\tau}_N$  and  $\bar{\tau}_R$  are mean relaxation times for momentum-conserving (normal) and momentum-

destroying (resistive) phonon scattering processes. The crucial part of (1.1) is the second inequality which requires a very pure crystal with high values of the thermal conductivity near its maximum. Up to now second sound has been detected in solid  $^4\text{He}$ ,  $^1,2$  solid  $^3\text{He}$ ,  $3,4$  NaF,  $5-8$  and in Bi.<sup>9</sup> In all these experiments essentially the same technique was used: A heat pulse is generated at one end of the crystal at  $t=0$  and the deviation of the local

temperature from the equilibrium value  $T_0$  is measured at the opposite side of the sample as a function of time for  $t > 0$ . The main features of the results are the following.

At very low temperatures  $T_0$  where there are virtually no phonon collisions except for boundary scattering, i. e.,

$$\bar{\Omega}\bar{\tau} \gg 1 \quad (1.2)$$

with

$$\bar{\tau}^{-1} = \bar{\tau}_N^{-1} + \bar{\tau}_R^{-1}, \quad (1.3)$$

two sharp pulses are seen corresponding to single longitudinal and transverse ballistic phonons traveling almost freely through the crystal with the transverse and longitudinal sound velocities  $C_t$  and  $C_l$ , respectively. (In an off-symmetry direction, where the transverse branches are not degenerate, two such pulses can be resolved.) When  $T_0$  is raised  $\bar{\tau}$  gets smaller. The longitudinal pulse (which arrives first since  $C_l > C_t$ ) normally does not change very much in shape; its amplitude, however, is diminished by the increasing number of phonon collisions. The behavior of the transverse pulse depends on the purity of the sample. For a dirty crystal its energy is also diminished and a broad diffusive signal with no definite arrival time appears which finally carries the whole intensity of the heat pulse. If the crystal is of a better quality a new feature arises: A secondary peak which is rather broad appears between the original transverse pulse of decreasing intensity and the diffusive ramp. This peak is identified with second sound and its velocity corresponds roughly to the theoretical value  $C_{II}$  which is given in (4.7). In a very good crystal the second sound shows more variation. The transverse pulse first broadens and shifts to later arrival times. For higher  $T_0$  this peak narrows again and is now identified with second sound with a velocity higher than  $C_{II}$ . The original transverse signal appears again at the slope of the second-sound pulse, now strongly damped. The second-sound peak now gradually moves to later arrival times and finally again disappears in favor of diffusive heat conduction.

The theoretical approach to these phenomena usually involves a phonon Boltzmann equation which determines the time behavior of the number of phonons carrying heat in the crystal. Such a collision equation has been derived from an anharmonic lattice Hamiltonian by various authors using equilibrium<sup>10-12</sup> or nonequilibrium Green's functions.<sup>13,14</sup> Whereas these derivations are involved, but in some sense straightforward, there is so far no explicit solution available for the Boltzmann equation. Sussmann and Thellung<sup>15</sup> have replaced the collision integral by relaxation times to find expressions for macroscopic quantities such

as energy density, etc., valid for  $\bar{\Omega}\bar{\tau} \ll 1$ . Thus they arrive at explicit expressions for velocity and damping of second sound. Meier<sup>16</sup> has generalized this procedure to an energy- and momentum-dependent relaxation rate. Guyer and Krumhansl<sup>17</sup> provided a more rigorous basis for discussing second sound in solids expanding the unknown phonon density in terms of the eigenfunctions of the collision operator  $C$ . Weiss<sup>18</sup> and Thellung and Weiss<sup>19</sup> have calculated explicit expressions for the attenuation of second sound by using only those eigenfunctions of  $C$  belonging to eigenvalue zero, a procedure which can be justified for small values of  $\bar{\Omega}\bar{\tau}$ . All the above-mentioned calculations, however, have yielded explicit results only for the hydrodynamic regime  $\bar{\Omega}\bar{\tau} \ll 1$ .

There are two attempts to cover the whole range  $\bar{\Omega}\bar{\tau} \gg 1$  to  $\bar{\Omega}\bar{\tau} \ll 1$  theoretically. Rogers<sup>8</sup> has used hydrodynamic equations for a single-branch model introducing a second viscosity depending strongly on  $\bar{\Omega}\bar{\tau}$ . Thus he obtained a continuous transition between the ballistic and hydrodynamic regimes describing the above-mentioned situation for a very pure crystal quite adequately. Recently Ranninger<sup>20</sup> treated the same problem in a semiphenomenological way using conservation laws for energy and momentum density. He also finds a smooth transition from transverse ballistic heat pulses to second sound, pointing out that the ballistic pulses are in fact also collective excitations of the phonon system. He furthermore pointed out that the second-sound velocity will decrease as a function of  $T_0$  due to the rising influence of resistive processes, a fact which has been observed in NaF as mentioned before. We shall compare Ranninger's conclusions to our results in Secs. IV and VI.

The purpose of the present work is to present explicit numerical results describing heat-pulse propagation in a dielectric crystal from  $\bar{\Omega}\bar{\tau} \gg 1$  to  $\bar{\Omega}\bar{\tau} \ll 1$ . The calculations are done for various strengths of resistive scattering, simulating crystals of different purity. The whole procedure is based on the linearized phonon Boltzmann equation for the deviation of the phonon density from its equilibrium value  $N_0$ , exposed in Sec. II. The local temperature  $T(\vec{r}, t)$  is defined in the usual way by the help of the deviation of the local energy from the equilibrium value. Section III is devoted to the solution of the Boltzmann equation approximating the normal and resistive parts of  $C$  by relaxation times  $\tau_N$  and  $\tau_R$ , taking into account, however, the zero eigenvalues (collision invariants!). We use an isotropic three-branch Debye model for the phonon spectrum in order to be able to calculate all expressions explicitly in terms of  $\tau_N$  and  $\tau_R$ . In Sec. IV the two limiting cases  $\bar{\Omega}\bar{\tau} \gg 1$  and  $\bar{\Omega}\bar{\tau} \ll 1$  are discussed analytically. The main results of

our analysis which are found numerically are presented in Sec. V and finally summarized in Sec. VI.

## II. MATHEMATICAL DESCRIPTION OF HEAT-PULSE EXPERIMENTS

The usual experimental arrangement for observing heat-pulse propagation consists of four parts: the sample through which the heat has to propagate; the heater, generating a short heat pulse at  $t=0$  on one side of the sample; a detector measuring the temperature on the opposite side as a function of  $t$ ; and a cryostat fixing the equilibrium temperature for  $t < 0$ . The dielectric sample crystal is assumed to be described by the usual phonon Hamiltonian

$$H = \sum_k \omega(k) (a_k^\dagger a_k + \frac{1}{2}) + \sum_{kk'k''} V_3(k, k', k'') (a_k + a_k^\dagger) \times (a_{k'} + a_{k'}^\dagger) (a_{k''} + a_{k''}^\dagger). \quad (2.1)$$

The anharmonic part  $H_{\text{anh}}$  involving  $V_3$  gives rise to phonon-phonon interactions. Higher anharmonicities could in principle be included without modifying the following arguments, at least not qualitatively. The sum over  $k = (\vec{k}, \lambda)$  involves a summation over wave vectors  $\vec{k}$  in the first Brillouin zone and over polarizations  $\lambda$ . For the analysis of heat-pulse propagation in the crystal we assume that all necessary information is contained in a space- and time-dependent phonon density  $N(\vec{k}, \vec{r}, t)$ . For  $t < 0$  the sample is in equilibrium with the surrounding heat bath, thus  $N$  does not depend on  $\vec{r}$  and  $t$  and is given by

$$N(k, \vec{r}, t < 0) = N_0(k) = (e^{\beta_0 \omega(k)} - 1)^{-1}, \quad (2.2)$$

with

$$\beta_0 = (k_B T_0)^{-1}, \quad (2.3)$$

neglecting hereby the fact that  $H_{\text{anh}}$  in (2.1) introduces a small correction to the energies  $\omega(k)$  and leads to phonons with finite lifetimes.

After the heat pulse, generated at  $t=0$ , the phonon distribution deviates from an equilibrium distribution. It is convenient to represent it as

$$N(k, \vec{r}, t > 0) = N_0(k) + m(k) \varphi(k, \vec{r}, t) \quad (2.4a)$$

with

$$m(k) = N_0(k) [N_0(k) + 1]. \quad (2.4b)$$

For low-intensity heat pulses the time dependence of  $\varphi$  is governed by a linearized transport equation. The latter has been derived from the Hamiltonian (2.1) by several authors (see Refs. 10, 12, 14, and others) using various mathematical techniques. Omitting the quasiparticle-interaction term included in the results of the above references and Fourier-Laplace transforming according to

$$\varphi(k, \vec{Q}, \Omega) = \int d\vec{r} e^{-i\vec{Q} \cdot \vec{r}} \int_0^\infty dt e^{-i\Omega t} \varphi(k, \vec{r}, t) \quad \text{for } \text{Im} \Omega \leq 0, \quad (2.5)$$

the phonon transport equation reads

$$[-i\Omega + i\vec{v}(k) \cdot \vec{Q} - C] \varphi(k, \vec{Q}, \Omega) = \varphi(k, \vec{Q}, t=0^+). \quad (2.6)$$

Here

$$\vec{v}(k) = \frac{\partial \omega(k)}{\partial \vec{k}} \quad (2.7)$$

is the phonon group velocity and  $C$  stands for the linearized Peierls-Boltzmann collision operator defined by

$$C\varphi(k, \vec{Q}, \Omega) = -\frac{\pi}{m(k)} \sum_{kk'k''} [m(k)m(k')m(k'')]^{1/2} \times \{2 |V_3(k, k', -k'')|^2 \delta(\omega(k) + \omega(k') - \omega(k'')) [\varphi(k, \vec{Q}, \Omega) + \varphi(k', \vec{Q}, \Omega) - \varphi(k'', \vec{Q}, \Omega)] \\ + |V_3(k, -k', -k'')|^2 \delta(\omega(k) - \omega(k') - \omega(k'')) [\varphi(k, \vec{Q}, \Omega) - \varphi(k', \vec{Q}, \Omega) - \varphi(k'', \vec{Q}, \Omega)]\},$$

where  $V_3$  is the anharmonic coupling parameter in the Hamiltonian (2.1).

Finding an appropriate form for the initial value  $\varphi(k, \vec{Q}, t=0^+)$  in (2.6), immediately after the short heat pulse has been applied, would in principle require detailed knowledge about the heating mechanism and the coupling between heater and sample. These questions have been examined by Kwok<sup>21</sup> and by von Gutfeld.<sup>22</sup> Here we assume the relaxation times of the heater to be very short. Thus a heater temperature  $T_h$  can be defined after the duration  $\Delta t$  of the pulse. If the coupling between heater and crystal is strong enough there will be a layer  $L$  in

the crystal where the local temperature has already adjusted to  $T_h$ . The thickness of  $L$  will be of the order of  $\Delta t \bar{c}$ ,  $\bar{c}$  being a mean phonon velocity of the crystal. Therefore

$$N(k, \vec{r}, t = \Delta t) \approx (e^{\beta(\vec{r}) \omega(k)} - 1)^{-1}, \quad (2.8)$$

where  $\beta(\vec{r}) = (k_B T_h)^{-1}$  for  $\vec{r}$  in the layer  $L$  and  $\beta(\vec{r}) = (k_B T_0)^{-1}$  for  $\vec{r}$  in the rest of the sample. For low-intensity pulses  $(T_h - T_0)/T_0$  is small, and (2.8) can be expanded:

$$N(k, \vec{r}, t = \Delta t) = N_0(k) - m(k) \omega(k) \Delta \beta_0(\vec{r}) \quad (2.9)$$

with

$$\Delta\beta_0 = \beta(\vec{r}) - \beta_0 \approx -\Delta T_0(\vec{r})/k_B T_0^2 \quad (2.10)$$

and

$$\varphi(k, \vec{Q}, t=0^+) = \omega(k) \Delta T_0(\vec{Q})/k_B T_0^2 = \omega(k) \Delta\beta_0(\vec{Q}) . \quad (2.11)$$

To calculate the temperature measured by the detector on the opposite sample boundary in the heat-pulse experiment we proceed as follows: We compute the local energy density

$$E(\vec{Q}, \Omega) = \sum_k \omega(k) N(k, \vec{Q}, \Omega) . \quad (2.12)$$

The deviation  $\Delta E$  from the equilibrium value  $E_0$ , given by

$$\Delta E(\vec{Q}, \Omega) = \sum_k \omega(k) m(k) \varphi(k, \vec{Q}, \Omega) , \quad (2.13)$$

is proportional to the deviation  $\Delta T$  of the local temperature:

$$\Delta E(\vec{Q}, \Omega) = C_v \Delta T(\vec{Q}, \Omega) , \quad (2.14)$$

$C_v$  being the heat capacity per unit volume. This is the usual definition of  $\Delta T$  (see, e.g., Refs. 10, 12, and 13). Assuming that the detectors used in these experiments can immediately adjust to the local temperature at the sample end the energy density (2.13) which can be calculated provided the solution  $\varphi(k, \vec{Q}, \Omega)$  to the transport equation (2.6) is known is a direct measure for the detected signal. These assumptions about the interaction between heater and sample, as well as sample and thermometer, are of course very much simplified and according to the experience of the experimentalists<sup>23</sup> the physical reality may be much more complicated.

### III. SOLUTION OF THE PHONON BOLTZMANN EQUATION IN COLLISION-TIME APPROXIMATION

Unfortunately it has not been possible so far to solve Eq. (2.6) exactly. A formal solution can be found by using the fact that the collision operator  $C$  is negative and symmetric in a Hilbert space of functions  $f(k)$  with the scalar product

$$\langle f_1, f_2 \rangle = \sum_k f_1(k) m(k) f_2(k) , \quad (3.1)$$

the weight function  $m$  being defined by (2.4b). Thus the phonon distribution function  $\varphi$  can be expanded with respect to the eigenfunctions  $\chi_\mu(k)$  of  $C$ :

$$\varphi(k, \vec{Q}, \Omega) = \sum_\mu a_\mu(\vec{Q}, \Omega) \chi_\mu(k) . \quad (3.2)$$

In the absence of umklapp processes there are four eigenfunctions with eigenvalue zero (collision invariants):

$$\chi_0(k) = \alpha_0 \omega(k) \equiv \hat{\omega}(k) \quad (3.3)$$

and

$$\chi_i(k) = \alpha_i k_i \equiv \hat{k}_i \quad (i = 1, 2, 3) \quad (3.4)$$

corresponding to the energy and the three Cartesian components of the wave vector  $\vec{k}$  of the phonons. The numbers  $\alpha_0$  and  $\alpha_i$  assure the proper normalization of the eigenfunctions. If momentum-destruction processes can take place  $\chi_0$  is the only collision invariant. All the remaining eigenvalues are negative. (For an infinite system the continuous spectrum of  $C$  extends, however, up to zero, see, e.g., Ref. 24.) These facts and the expansion (3.2) have been used to derive equations describing second sound, thermal conductivity, and the elastic behavior, etc., in the limit of small  $\vec{Q}$  and  $\Omega$  by various authors.<sup>10,13,17</sup> For arbitrary values of  $\vec{Q}$  and  $\Omega$ , however, Eq. (2.6) would have to be solved by some numerical procedure. Although this may yield interesting information about the detailed behavior of the experimental quantities, as it has been shown by Maris<sup>25</sup> and by Meier and Beck<sup>26</sup> for the velocity of sound in superfluid helium, a simple and intuitive collision-time approximation for  $C$  still seems to be a good method to describe the qualitative behavior of the phonon system. This approximation is introduced in the following way: The expansion (3.2) is written in the form

$$\varphi(k, \vec{Q}, \Omega) = a_0(\vec{Q}, \Omega) \hat{\omega}(k) + \sum_{i=1}^3 a_i(\vec{Q}, \Omega) \hat{k}_i + \chi(k, \vec{Q}, \Omega) , \quad (3.5)$$

$\chi$  standing for all the terms involving eigenfunctions with nonzero eigenvalues.  $C$  is split into a normal part  $C_N$  and a resistive part  $C_R$ , the latter describing momentum-nonconserving processes:

$$C = C_N + C_R . \quad (3.6)$$

The effect of  $C$  operating on the right-hand side of (3.5) is approximated by

$$\begin{aligned} C\hat{\omega}(k) &= 0 , \\ C\hat{k}_i &= C_R \hat{k}_i = -\frac{1}{\tau_R} \hat{k}_i , \\ C\chi &= -\left(\frac{1}{\tau_N} + \frac{1}{\tau_R}\right) \chi = -\frac{1}{\tau} \chi . \end{aligned} \quad (3.7)$$

$\tau_N$  and  $\tau_R$  are relaxation times for normal and resistive processes, respectively. In principle they may be chosen to be  $\vec{k}$  and  $\lambda$  dependent. The representation (3.7) for  $C$  seems to be appropriate for the case where umklapp processes are still less frequent than normal ones. If both types of collisions are at least of equal importance ( $\tau_R/\tau_N \lesssim 1$ ) it is more adequate to use only one collision invariant,  $\hat{\omega}(k)$ , and to replace (3.5) and (3.7) by

$$\varphi(k, \vec{Q}, \Omega) = a_0(\vec{Q}, \Omega) \hat{\omega}(k) + \chi'(k, \vec{Q}, \Omega) \quad (3.5')$$

and

$$C\chi'(k, \vec{Q}, \Omega) = -(1/\tau) \chi'(k, \vec{Q}, \Omega) . \quad (3.7')$$

The results for this case, which is actually less interesting since there will be no second sound, will be listed at the end of this section for the sake

of comparison.

By means of (3.7) the contribution  $\chi$  to the solu-

tion (3.5) of our Boltzmann equation can be represented as

$$\chi(k, \vec{Q}, \Omega) = D^{-1}(k, \vec{Q}, \Omega) \left[ \varphi(k, \vec{Q}, t=0^+) - [-i\Omega + i\vec{Q} \cdot \vec{v}(k)] a_0(\vec{Q}, \Omega) \hat{\omega}(k) - \left( -i\Omega + i\vec{Q} \cdot \vec{v}(k) + \frac{1}{\tau_R} \right) \sum_i a_i(\vec{Q}, \Omega) \hat{k}_i \right], \quad (3.8)$$

with

$$D(k, \vec{Q}, \Omega) = -i\Omega + i\vec{Q} \cdot \vec{v}(k) + 1/\tau. \quad (3.9)$$

The approximations described here correspond to the model solution of the Boltzmann equation valid for a gas of particles discussed by Foch and Ford.<sup>27</sup> We shall use their method of analyzing the results of such a calculation in Sec. V.

The unknown expansion coefficients  $a_0$  and  $a_i$  are determined by the help of the conservation laws for energy and quasimomentum. These can be derived by multiplying (2.6) by  $\hat{\omega}$  and  $\hat{k}_i$ , respectively, and taking the scalar product as defined in (3.1):

$$-i\Omega a_0 + \sum_i Q_i \langle \hat{\omega}, v_i \varphi \rangle = \langle \hat{\omega}, \varphi(t=0^+) \rangle \quad (3.10)$$

and

$$-i\Omega a_i + i \sum_l \Omega_l \langle \hat{k}_i, v_l \varphi \rangle + \langle \hat{k}_i, (1/\tau_R) \varphi \rangle = \langle \hat{k}_i, \varphi(t=0^+) \rangle. \quad (3.11)$$

Here the symmetry of  $C$  and the relations (3.5) and (3.7) have been used, and  $v_i$  are the Cartesian components of the group velocity (2.7).

So far our expressions are valid for arbitrary phonon spectra  $\omega(\vec{k}, \lambda)$ . To be able to evaluate (3.10) and (3.11) by analytical methods as far as possible we now introduce an isotropic Debye model with two transverse and one longitudinal phonon branches, i. e.,

$$\omega(\vec{k}, \lambda) = C_\lambda |\vec{k}|, \quad \lambda = 1, 2, 3. \quad (3.12)$$

Furthermore we replace the relaxation times  $\tau_R(\vec{k}, \lambda)$  and  $\tau_N(\vec{k}, \lambda)$  entering the scalar products in (3.10) and (3.11) by mean values  $\tau_{N,\lambda}$  and  $\tau_{R,\lambda}$  appropriate for thermal phonons. In an isotropic system the vector  $\vec{a}(\vec{Q}, \Omega)$  can be split into a transverse part  $\vec{a}^T$  and a contribution parallel to  $\vec{Q}$ :

$$\vec{a}(\vec{Q}, \Omega) = b(\vec{Q}, \Omega) \vec{Q} / |\vec{Q}| + \vec{a}^T(\vec{Q}, \Omega). \quad (3.13)$$

It is easy to see that the energy density  $\Delta E$  which has finally to be calculated does not couple to  $\vec{a}^T$ . Therefore, we end up with an inhomogeneous system of two linear equations for the quantities  $a_0$  and  $b$ :

$$(\Omega + C_{11})a_0 + C_{12}b = e_1 \Delta \beta_0, \quad (3.14)$$

$$C_{21}a_0 + (\Omega + C_{22})b = e_2 \Delta \beta_0. \quad (3.15)$$

The intermediate calculations are somewhat complicated but straightforward. Observing the rela-

tion

$$\int_0^{k_{\max}} dk k^4 m(\vec{k}, \lambda) \approx \int_0^\infty dk k^4 m(k, \lambda) = \frac{1}{\beta_0^5 C_\lambda^5} \int_0^\infty dx x^4 \frac{e^x}{(e^x - 1)^2} = \frac{1}{\beta_0^5 C_\lambda^5} \gamma \quad (3.16)$$

and the definition

$$S_n \equiv \sum_\lambda \frac{1}{C_\lambda^n}, \quad (3.17)$$

the coefficients  $C_{ij}$  and  $e_i$  can be written as

$$\begin{aligned} C_{11} &= \frac{iQ}{2S_3} \sum_\lambda \frac{1}{C_\lambda^2 \tau_\lambda} I_1(\lambda), \\ C_{12} &= \frac{iQ}{2} \left( \frac{3}{S_3 S_5} \right)^{1/2} \sum_\lambda \frac{1}{C_\lambda^3 \tau_{N,\lambda}} I_2(\lambda), \\ C_{21} &= \frac{iQ}{2} \left( \frac{3}{S_3 S_5} \right)^{1/2} \sum_\lambda \frac{1}{C_\lambda^3 \tau_\lambda} I_2(\lambda), \\ C_{22} &= \frac{3iQ}{2S_5} \sum_\lambda \frac{1}{C_\lambda^4 \tau_{N,\lambda}} I_3(\lambda) + \frac{i}{3} \sum_\lambda \frac{1}{\tau_{R,\lambda}}, \\ e_1 &= -i \left( \frac{\pi\gamma}{\beta_0^5 S_3} \right)^{1/2} \sum_\lambda \frac{\Omega + i/\tau_\lambda}{C_\lambda^3} I_0(\lambda), \\ e_2 &= -iQ \left( \frac{3\pi\gamma}{\beta_0^5 S_5} \right)^{1/2} \sum_\lambda \frac{1}{C_\lambda^3} I_2(\lambda), \end{aligned} \quad (3.18)$$

with

$$\tau_\lambda^{-1} = \tau_{N,\lambda}^{-1} + \tau_{R,\lambda}^{-1}$$

and

$$I_n(\lambda) = \int_{-1}^{+1} dz \frac{z^n}{Q C_\lambda z - \Omega - i/\tau_\lambda}. \quad (3.20)$$

According to (2.13) and (3.3) the local temperature  $\Delta T$  introduced in (2.14) is closely related to the coefficient  $a_0$ :

$$\Delta T = a_0 / \alpha_4 C_v. \quad (3.21)$$

It is proportional to the initial temperature deviation  $\Delta T_0$ :

$$\Delta T(\vec{Q}, \Omega) = R(\vec{Q}, \Omega) \Delta T_0(\vec{Q}), \quad (3.22)$$

where the response function  $R$  is given by

$$R = \frac{(\Omega + C_{22})f_1 + C_{12}f_2}{(\Omega + C_{11})(\Omega + C_{22}) - C_{12}C_{21}} \quad (3.23)$$

with

$$f_1 = -i \frac{1}{2S_3} \sum_{\lambda} \frac{\Omega + i/\tau_{\lambda}}{C_{\lambda}^3} I_0(\lambda),$$

$$f_2 = iQ \frac{1}{2} \left( \frac{3}{S_3 S_5} \right)^{1/2} \sum_{\lambda} \frac{1}{C_{\lambda}^3} I_2(\lambda), \quad (3.24)$$

the specific heat  $C_v$  for this model being

$$C_v = 4\pi\gamma S_3 k_B^4 T_0^3 (\hbar=1). \quad (3.25)$$

Had we used the formulas (3.5') and (3.7') for the case of strong resistive processes we would have found

$$\Delta T(\vec{Q}, \Omega) = R'(\vec{Q}, \Omega) \Delta T_0(\vec{Q}) \quad (3.26)$$

with

$$R'(\vec{Q}, \Omega) = \frac{f_1(\vec{Q}, \Omega)}{\Omega + C_{11}(\vec{Q}, \Omega)}. \quad (3.27)$$

This result has to be used if  $\tau_R/\tau_N \leq 1$ .

#### IV. CALCULATION OF $\Delta T(\vec{Q}, \Omega)$ FOR THE HYDRODYNAMIC AND THE BALLISTIC REGIME

The time behavior of  $\Delta T$  for  $t > 0$  is given by

$$\Delta T(\vec{Q}, t) = \int d\Omega e^{-i\Omega t} R(\vec{Q}, \Omega) \Delta T_0(\vec{Q}). \quad (4.1)$$

If  $R^{-1}(\vec{Q}, \Omega)$  were an analytic function of  $\Omega$  for  $\text{Im}\Omega < 0$  the possible excitations of the system would be determined by its zeros in the lower-half of the complex frequency plane. The coefficients  $C_{ij}$  and  $f_i$  entering  $R$  have, however, branch cuts for

$$-QC_{\lambda} \leq \text{Re}\Omega \leq QC_{\lambda}$$

and

$$\text{Im}\Omega = -i/\tau_{\lambda} \quad (4.2)$$

since they involve integrals of the form (3.20). This nonanalyticity will be essential for discussing the general behavior of the response function in Sec. V. Here we want to treat two limiting cases, namely  $\bar{\Omega} \bar{\tau} \gg 1$  and  $\bar{\Omega} \bar{\tau} \ll 1$ , where  $\bar{\Omega}$  is a characteristic frequency of the heat pulse being of the order of the inverse of the pulse duration  $\Delta t$  and  $\bar{\tau}$  represents a mean relaxation time of the thermal phonons. In both cases  $R$  can be approximated by a simple analytic function.

##### A. $\bar{\Omega} \bar{\tau} \gg 1$ , Collisionless Domain

Here the coefficients  $C_{ij}$  are small compared to the relevant values of  $\Omega$ , since they are all pro-

portional to  $\bar{\tau}_{\lambda}$ ; therefore the denominator in (3.23) can be replaced by  $\Omega^2$  and

$$R(\vec{Q}, \Omega) = \frac{-i}{2S_3} \sum_{\lambda} \frac{1}{C_{\lambda}^3} \int dz \frac{1}{QC_{\lambda}z - \Omega - i/\tau_{\lambda}}$$

$$= \frac{1}{2iS_3} \sum_{\lambda} \frac{1}{QC_{\lambda}^4} \ln \frac{\Omega - QC_{\lambda} + i/\tau_{\lambda}}{\Omega + QC_{\lambda} + i/\tau_{\lambda}}. \quad (4.3)$$

This is exactly the same result one would obtain by simply replacing the collision operator  $C$  in (2.6) by the inverse relaxation time  $1/\tau_{\lambda}$  without taking into account the collision invariants by the prescriptions (3.7). In this regime where there are only few collisions during a period of oscillation  $1/\Omega$  heat propagates in the form of single ballistic phonons with velocity  $C_{\lambda}$  and lifetime  $\tau_{\lambda}$  [if the phonon spectrum were not linear as assumed in (3.14),  $C_{\lambda}$  would be replaced by an average group velocity of thermal phonons]. The total effect of these phonons to the heat pulse is calculated by integrating over all angles  $\theta$  between the propagation direction  $\vec{Q}$  of the pulse and the wave vector  $\vec{K}$  of the individual phonons ( $z = \cos\theta$ ) which leads to the log-type behavior of  $R$  in (4.3). The relative weight  $g_{\lambda}$  of the three polarizations is given by

$$g_{\lambda} = 1/C_{\lambda}^3. \quad (4.4)$$

Since  $C_i > C_t$ , the transverse branches  $t_1$  and  $t_2$  are stronger than the longitudinal ones. This effect is most pronounced in those symmetry directions where the transverse velocities are degenerate. Here one intense transverse pulse and a weak longitudinal signal are observed; see, e.g., Refs. 5, 7, and 9. Since collisions are unimportant in this domain it is not further surprising that the form (3.26) involving  $R'$ , appropriate for  $\tau_R/\tau_N \leq 1$ , leads to the result (4.3) too, in the regime  $\bar{\Omega} \bar{\tau} \gg 1$ .

##### B. $\bar{\Omega} \bar{\tau} \ll 1$ , Hydrodynamic Regime

In this case the integrand in (3.20) can be expanded:

$$(QC_{\lambda}z - \Omega - i/\tau_{\lambda})^{-1} \approx i\tau_{\lambda} [1 - i\tau_{\lambda}(QC_{\lambda}z - \Omega) + \dots], \quad (4.5)$$

neglecting higher-order terms, since they are small for the representative values of  $\Omega$ . In this limit  $R(\vec{Q}, \Omega)$  reads

$$R(\vec{Q}, \Omega) \approx i \frac{\Omega + i/\tau_R}{(\Omega + iQ^2C_{11}^2\tau)[\Omega + i/\tau_R + i\frac{Q^2}{2}C_{11}^2\tau(1-\alpha)] - Q^2C_{11}(1-\alpha)(1+2i\Omega\tau)}, \quad (4.6)$$

where the following definitions have been introduced:

$$C_{11}^2 = S_3/3S_5, \quad (4.7)$$

$$C_{11}^{\prime 2} = S_1/3S_3, \quad (4.8)$$

$$\alpha = \tau/\tau_R. \quad (4.9)$$

Furthermore, for simplicity, we have introduced

mean relaxation times  $\tau_R$ ,  $\tau_N$ , and  $\tau$  representing averages of  $\tau_{N,\lambda}$ ,  $\tau_{R,\lambda}$ , and  $\tau_\lambda$  over polarizations. The possible excitations are given by the zeros of the denominator

$$\Omega^2 + i\Omega(aQ^2\tau + 1/\tau_R) - Q^2d^2 = 0 + O(\bar{\Omega}\bar{\tau})^2, \quad (4.10)$$

with

$$\begin{aligned} a &= C_{II}'^2 - \frac{1}{5}C_{II}^2(1 - \alpha), \\ d^2 &= C_{II}^2 + \alpha(C_{II}'^2 - C_{II}^2). \end{aligned} \quad (4.11)$$

They are situated at

$$\Omega_\pm = -\frac{1}{2}i(aQ^2\tau + 1/\tau_R) \pm QC_2 \quad (4.12)$$

with

$$C_2 = C_{II} \left[ 1 - \frac{9\alpha}{10} + \frac{\alpha}{2} \left( \frac{C_{II}'}{C_{II}} \right)^2 - \frac{1}{4Q^2C_{II}^2\tau_R^2} \right]^{1/2}. \quad (4.13)$$

For  $QC_{II}\tau_R \gtrsim 1$  they describe a collective mode of heat propagation—second sound. Since

$$\alpha = \tau/\tau_R = (1 + \tau_R/\tau_N)^{-1}$$

and  $1/\tau_R$  itself are increasing functions of the ambient temperature  $T_0$  as soon as umklapp processes tend to increase more than normal processes (see Sec. V for explicit  $T$  dependence of  $\tau_R$ ,  $\tau_N$ ) the velocity of second sound will decrease from its “classical” value  $C_{II}$ , which is valid for  $\tau_R \rightarrow \infty$ , and will finally go to zero when  $QC_{II}\tau_R \lesssim 2$ . This fact is observed experimentally and has also been accounted for by Ranninger<sup>20</sup> who finds

$$C_2 \approx \pm C_{II}/(1 + 2\alpha)^{1/2}. \quad (4.14)$$

We believe, however, that in the window range where second sound is observable, i.e., for

$$\bar{\Omega}\bar{\tau} > 1 > \bar{\Omega}\bar{\tau}_N, \quad (4.15)$$

the quantity  $\alpha = (1 + \tau_R/\tau_N)^{-1}$  should still be much smaller than 1, in order that (4.15) be fulfilled. Therefore the variation of  $C_2$  according to (4.14) seems to be weaker than that of (4.13) where the term  $1/4Q^2C_{II}^2\tau_R^2$  will finally drive  $C_2$  to zero, when  $QC_{II}\tau_R$  becomes small enough. In this domain the momentum-destroying collisions prohibit the wave-like propagation of a heat pulse. Heat is transported in a purely diffusive way. If  $\tau_R \ll \tau_N$  the solutions  $\Omega_\pm$  in (4.13) can be approximated by putting  $\alpha = 1$  and  $\tau_R = \tau$  which yields

$$\Omega_\pm \approx -iQ^2C_{II}'^2\tau \approx 0, \quad (4.16)$$

and

$$\Omega_- \approx -i/\tau. \quad (4.17)$$

Had we performed the hydrodynamic limit in  $R'$ , Eq. (3.27), which is valid for  $\tau_R < \tau_N$ , we would have found

$$R'(\bar{Q}, \Omega) \approx i/(\Omega + iQ^2C_{II}'^2\tau). \quad (4.18)$$

The pole of this expression, situated at  $\Omega = -iQ^2C_{II}'^2\tau$  and representing ordinary heat conduction, thus coincides with the limit  $\Omega_+$  in (4.16) for  $\tau_R \ll \tau_N$ .

It is interesting to note that in the two limiting situations Secs. IV A and IV B the response of the system is determined by different contributions of  $R$ : for  $\bar{\Omega}\bar{\tau} \gg 1$  the denominator reduces to  $\Omega^2$  whereas the log terms in the numerator yield the ballistic *single-particle-like* excitations. In the opposite case ( $\bar{\Omega}\bar{\tau} \ll 1$ ) the numerator behaves regularly in the important domain of the complex variable  $\Omega$  whereas the denominator produces zeros describing *collective* phenomena such as heat conduction and second sound. These facts strongly suggest that ballistic heat pulses and second-sound waves are in some sense two completely distinct phenomena between which there is no smooth transition. This will become even more apparent in the analysis of Sec. V.

#### V. NUMERICAL ANALYSIS OF THE RESPONSE FUNCTION

The final goal would of course be to calculate the temperature response  $\Delta T(\bar{r}, t)$  for points  $\bar{r}$  at the sample end, i.e., to evaluate the integral

$$\Delta T(\bar{r}, t) = \int d^3Q e^{i\bar{Q} \cdot \bar{r}} \int d\Omega e^{-i\Omega t} R(\bar{Q}, \Omega) \Delta T_0(\bar{Q}). \quad (5.1)$$

It would require extensive numerical work to obtain reliable results and the numerical procedure would probably hide the mathematical facts which are important to understand the transition from the ballistic to the hydrodynamic regime. We therefore propose two different methods of investigating the properties of  $R$  and  $R'$ , respectively: (a)  $R(\bar{Q}, \Omega)$  is considered as a function of the complex variable  $\Omega$ , for fixed  $\bar{Q}$ . The behavior of its singularities is studied as they depend on different magnitudes of  $\tau$ ,  $\tau_R$ , and  $\tau_N$ . (b)  $R(\bar{Q}, \Omega)$  is calculated as a function of the real variable  $\Omega$  for a fixed, representative value of  $\bar{Q}$ . The results of (a) and (b) should be sufficient to interpret the general behavior of (5.1) and to compare these results with the experimental data which show  $\Delta T(\bar{r}, t)$  for  $|\bar{r}| = L$ ,  $L$  being the sample length.

Let us first introduce dimensionless variables by representing the sound velocities  $C_\lambda$  and the wave vectors  $\bar{Q}$  as multiples of the lower transverse velocity  $C_0$  and a representative wave vector  $Q_0$ , respectively:

$$C_\lambda = d_\lambda C_0, \quad (5.2a)$$

$$Q = q Q_0. \quad (5.2b)$$

$Q_0$  can be related to a characteristic frequency  $\Omega_0$  of the heat pulse, given by  $\Omega_0 = 2\pi/\Delta t$ , by

$$Q_0 \approx \Omega_0/C_0. \quad (5.3)$$

Defining a dimensionless quantity  $x$  by

$$\Omega = xqC_0Q_0 = xC_0Q \quad (5.4)$$

and dimensionless relaxation coefficients  $\xi$ ,  $\xi_N$ , and  $\xi_R$  by

$$\frac{1}{\tau_N} = \frac{Q_0C_0}{\xi_N} = \frac{\Omega_0}{\xi_N}, \quad (5.5)$$

etc., the results of Sec. III can be written as

$$R(q, x) = \frac{i}{2D_3\Omega_0q} \frac{(x+a_{22})h_1 + a_{12}h_2}{(x+a_{11})(x+a_{22}) - a_{12}a_{21}} \\ \equiv \frac{i}{2D_3\Omega_0q} \frac{z(q, x)}{D(q, x)} \quad (5.6)$$

and

$$R'(q, x) = \frac{i}{2D_3\Omega_0q} \frac{h_1}{x+a_{11}} \equiv \frac{i}{2D_3\Omega_0q} \frac{z'(q, x)}{D'(q, x)}. \quad (5.7)$$

Here

$$D_n = \sum_{\lambda} d_{\lambda}^n \quad (5.8)$$

and the coefficients  $a_{ij}$  and  $h_i$  follow from  $C_{ij}$  [(3.18)] and  $f_i$  [(3.24)] by going to the new variables defined in (5.2)–(5.5). It can immediately be seen that, besides the factor  $i/2D_3\Omega_0q$ , the response functions  $R$  and  $R'$  depend on  $q$  only via the quantities  $\xi q$ ,  $\xi_N q$ , and  $\xi_R q$ . Therefore we can fix  $q$  without losing generality since a variation of  $q$  amounts to a new scaling of  $\xi$ ,  $\xi_N$ , and  $\xi_R$ . We chose  $q=1$ , thus our results should be representative for  $\bar{Q} \approx \bar{Q}_0$ , the main  $\bar{Q}$  vector given by (5.3).

Instead of treating  $\xi_N$  and  $\xi_R$  as free parameters in  $R$  and  $R'$  we define a temperature dependence for these relaxation times which can serve as a model for NaF crystals of different purities. We put

$$1/\xi_R = 1/\xi_u + 1/\xi_I \quad (5.9)$$

and

$$1/\xi_N = A_N T^5 \quad \text{for normal processes,} \quad (5.10)$$

$$1/\xi_u = A_u T^4 e^{-\Theta/3.5T} \quad \text{for umklapp processes,} \quad (5.11)$$

$$1/\xi_I = A_I T. \quad (5.12)$$

$A_N$  and  $A_u$  are considered to be intrinsic constants of these crystals, independent of the quality of the crystal, whereas  $\xi_I$  stands for all momentum-non-conserving processes other than phonon-phonon umklapp collisions, such as dislocations, impurities, etc. Although these processes may have a temperature dependence different from (5.12) the quantity  $A_I$  may serve as a parameter to distinguish in an easy way between different purities and growth qualities of the crystal. In accordance with the definition (5.5) and the numbers quoted in Ref. 7 we put

$$\Theta = 350^\circ \text{K},$$

$$A_N = 0.7 \times 10^{-5} ^\circ \text{K}^{-5}, \quad (5.13)$$

$$A_u = 0.2 \times 10^{-2} ^\circ \text{K}^{-4}, \quad (5.14)$$

whereas  $A_I$  varies between 0.0 and  $0.08 ^\circ \text{K}^{-1}$ . Now we can proceed to the analysis of  $R$  and  $R'$  for different values of  $T$  and  $A_I$ .

#### A. Analytic Properties of $R$ and $R'$

First we observe that the numerators  $z$  and  $z'$  are analytic functions, except for logarithmic singularities at

$$x = \pm d_{\lambda} - i/q\xi(T). \quad (5.15)$$

These singularities represent single ballistic phonons. They move away from the real axis, as the damping  $\xi^{-1}(T)$  increases; see Fig. 1. Furthermore it is easy to establish that  $z$  and  $z'$  go to zero for  $x \rightarrow -i/q\xi$ ; namely,

$$z(q, x) \propto (x + i/q\xi)^2 \quad \text{for } x + i/q\xi \rightarrow 0, \quad (5.16)$$

$$z'(q, x) \propto (x + i/q\xi) \quad \text{for } x + i/q\xi \rightarrow 0. \quad (5.17)$$

It is somewhat more difficult to analyze  $D$  and  $D'$ . First we remark that there are zeros for  $x = -i/q\xi$ ; namely,

$$D(q, x) \propto (x + i/q\xi)^2 \quad \text{for } x \rightarrow -i/q\xi, \quad (5.18)$$

$$D'(q, x) \propto (x + i/q\xi) \quad \text{for } x \rightarrow -i/q\xi. \quad (5.19)$$

These zeros of the denominators do not produce any singularities in  $R$  and  $R'$  since, according to (5.16) and (5.17), the numerators  $z$  and  $z'$  vanish with the same power of  $x + i/q\xi$ . Furthermore the coefficients  $a_{ij}$  also have logarithmic singularities at the values (5.15) of  $x$ . Since  $a_{ij} \propto 1/\xi q$  or  $1/\xi_N q$ , respectively, these singularities do not have a sig-

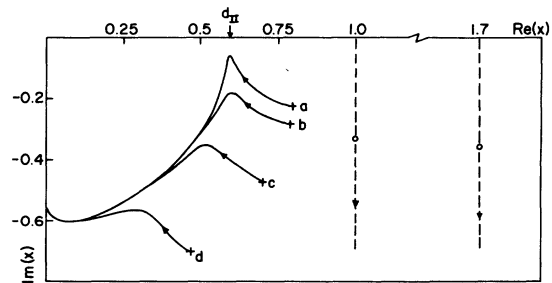


FIG. 1. Qualitative behavior of the different singularities of  $R$  in the complex  $x$  plane as functions of  $T$ . The arrows indicate the direction in which the positions of the singularities move for rising  $T$ . Dashed-circles, ballistic single-phonon excitations moving away from the real axis at constant velocities  $d_I=1$  and  $d_I=C_I/C_T=1.7$ ; crosses, incipient second-sound poles at  $T=T_c$ ; directed line, locus of the second-sound poles for  $T>T_c$ . The letters a–d give the corresponding value of  $A_I$ : (a)  $A_I=0$ , (b)  $A_I=0.01$ , (c)  $A_I=0.04$ , (d)  $A_I=0.08$ .



TABLE I. Critical values of the relaxation time for which second sound starts developing.

$A_I$ (°K <sup>-1</sup> )	$(q\xi_N)_c^{-1}$	$(q\xi_R)_c^{-1}$	$(q\xi)_c^{-1}$	$T_c^a$ (°K)	$d_2^b$
0.0	0.225	0	0.225	7.97	0.805
0.01	0.208	0.078	0.286	7.85	0.787
0.02	0.193	0.155	0.348	7.73	0.763
0.03	0.180	0.227	0.409	7.62	0.735
0.04	0.168	0.301	0.469	7.52	0.699
0.05	0.158	0.371	0.530	7.43	0.657
0.06	0.149	0.441	0.590	7.34	0.606
0.07	0.141	0.508	0.650	7.26	0.543
0.08	0.134	0.575	0.710	7.19	0.466

<sup>a</sup> $T$  value which produces the critical relaxation times in our model for  $q=1$ .

<sup>b</sup>Value of the velocity of incipient second sound in fraction of  $C_t$ . In our model  $C_l/C_t=1.7$ . Thus  $d_{II}=C_{II}/C_t=0.596$ . The attenuation of second sound at the onset is equal to  $(q\xi)_c^{-1}$ .

nificant influence, as long as  $(q\xi_N)^{-1}$  and  $(q\xi)^{-1}$  are small, and where  $(q\xi_N)^{-1}$  and  $(\xi q)^{-1}$  are larger the  $x$  values (5.15) are out of the physically interesting region, characterized by  $\text{Im}z$  being small. The crucial thing is, however, to look for possible zeros other than (5.18) and (5.19) of  $D$  and  $D'$ , which give rise to the collective excitations in  $R$  and  $R'$ . To do this we use the winding theorem for a complex function  $F$  which proved to be a useful tool in the model calculations of Ford and Foch.<sup>27</sup> It says that the number of zeros of  $F$  in a domain where  $F$  is analytic is equal to the number of times the graph of  $F$  encircles the origin of the  $F$  plane.<sup>28</sup> Because of the logarithmic singularities of  $D$  and  $D'$  we have to divide the  $x$  plane into two domains  $G_1$  and  $G_2$ :  $G_1$  encircled by the line  $\text{Im}x = -i/q\xi + i\epsilon$  and the corresponding upper semicircle at infinity, and  $G_2$  contoured by  $\text{Im}x = i/q\xi - i\epsilon$  and the lower semicircle. The result of this procedure is that there are critical values  $(\xi q)_c$  and  $(\xi_R q)_c$  such that there is no further zero than (5.18) and (5.19) in both domains,  $G_1$  and  $G_2$ , as long as  $\xi q > (\xi q)_c$  and  $\xi_R q > (\xi_R q)_c$ . For  $q\xi_R < (q\xi_R)_c$  and  $q\xi < (q\xi)_c$  there are, however, zeros in  $G_1$ ; namely, two second-sound poles with real parts of opposite sign for  $R(q, x)$  and a heat-conduction pole with  $\text{Re}x=0$  for  $R'(q, x)$ . Thus, in our model, there are characteristic values  $T_c$ , or  $(q\xi)_c$  and  $(q\xi_R)_c$ , such that the collective modes start to exist at  $T = T_c$  and develop for  $T > T_c$ , whereas they vanish for  $T < T_c$ . This peculiar fact is of course due to the nonanalytic behavior of  $R$  and  $R'$ , and has also been found in the model of Ref. 27. Some possible onset values  $T_c$ ,  $(\xi q)_c$ , and  $(\xi_R q)_c$  are listed in Table I, and the behavior of the poles for  $T > T_c$  is described in Fig. 1. Here we summarize some important facts.

(i) The onset value  $x_0$  of the second-sound pole

depends strongly on  $\xi_R$ . The smaller  $A_I$ , i.e., the purer the crystal, the smaller  $|\text{Im}x_0|$ , the damping, and the larger  $\text{Re}x_0$ , the velocity. For  $A_I=0$  the velocity lies almost half-inbetween  $C_{II}$  and  $C_t$ ! This explains the fact that in a pure crystal the transverse pulse seems to move continuously into the second-sound pulse (see, e.g., the pictures of Ref. 6), whereas for a crystal of lower quality, with higher  $A_I$ , where  $\text{Re}x_0 \approx d_{II}$ , the second-sound peak is broader and shows up as a distinct peak separated from the transverse one, as it can be seen in certain pictures of Ref. 7.

(ii) If  $A_I$  is not too large, the pole moves first toward the real axis, corresponding to a narrowing of the second-sound peak, and the velocity decreases approaching the value  $C_{II}$ .

(iii) As soon as  $T$  rises more, such that intrinsic umklapp processes become important, the velocity decreases further and the damping gets larger.

(iv) Finally, as  $\tau_R/\tau_N \leq 1$ , the poles reach the imaginary axis showing the same behavior as the heat conduction pole of  $R'$ .

(v) The values of  $(\xi q)_c$  show, that second sound can already develop for  $\bar{\Omega}\bar{\tau}_N \gtrsim 1$ , which suggests that the first inequality of the window condition may indeed be too strong.

#### B. Behavior of $R(q, x)$ and $R'(q, x)$ for Real $x$

In Sec. VA we have investigated the possible excitations of the phonon system in a weak-pulse experiment. One result was that ballistic pulses and the collective modes originate in two distinct kinds of singularities of  $R$  and  $R'$ . It may thus be interesting to plot these response functions, or at least their imaginary parts, as a function of the real frequency  $x$ , as they would enter the integral (5.1), to see how the different regimes match at  $\bar{\Omega}\bar{\tau} \approx 1$ . This is done in Fig. 2 for  $A_I=0.001$  °K<sup>-1</sup> showing  $\text{Im}R(q, x)$ , which can be interpreted as a spectral function for the heat pulse. It shows that, for fixed  $q$ , the transverse pulse, which is rather high and narrow at low  $T$ , broadens and is finally replaced by a new pulse, the second-sound pulse with a velocity  $\xi(T)$  behaving in the way described before. Since, for a short initial pulse, (5.7) contains an integral over a wide range of  $q$  values, it is possible that  $\Delta T(\vec{r}, t)$ , at an intermediate range  $\bar{\Omega}\bar{\tau} \gtrsim 1$ , shows both a transverse and second-sound pulse, at the same time, since the higher  $q$  values with  $q\xi > (\xi q)_c$  still contribute mainly to the ballistic peak, whereas smaller  $q$ 's with  $q\xi < (\xi q)_c$  already build up the second-sound peak.

#### VI. CONCLUSIONS AND SUMMARY

The phonon Boltzmann equation has been solved in a collision-time approximation including normal and resistive scattering for a Debye solid with two degenerate transverse and one longitudinal phonon

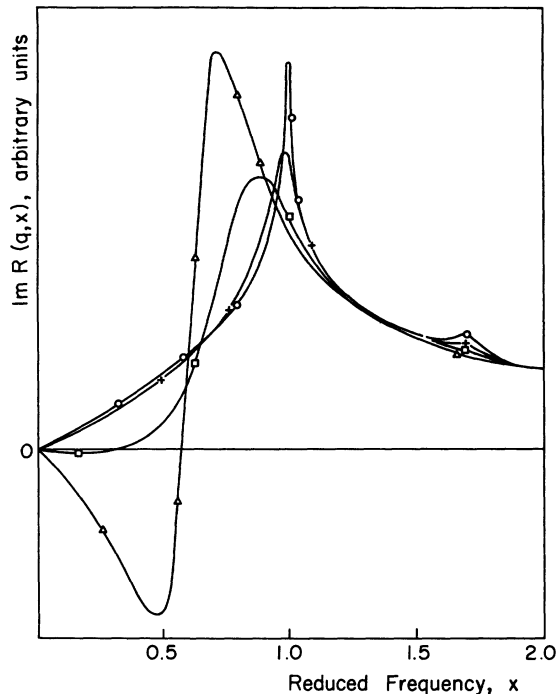


FIG. 2. Imaginary part of  $R(q, x)$ , Eq. (5.6), is plotted as a function of  $x$  for  $q=1$  and  $A_I=0.001$ . The curves for different temperatures—namely, circles, 2°K,  $(q\xi_N)^{-1}=0.0002$ ,  $(q\xi_R)^{-1}=0.002$ ; crosses, 5°K,  $(q\xi_N)^{-1}=0.022$ ,  $(q\xi_R)^{-1}=0.005$ ; squares, 8°K,  $(q\xi_N)^{-1}=0.229$ ,  $(q\xi_R)^{-1}=0.008$ ; triangles, 11°K,  $(q\xi_N)^{-1}=1.13$ ,  $(q\xi_R)^{-1}=0.074$ —exhibit the transition between the ballistic approximation (4.3) and the second-sound approximation (4.6) to  $R(q, x)$ . The longitudinal peak at  $x=1.7$  vanishes in the tail of the transverse, whereas the latter broadens, shifts to smaller  $x$  values and is finally replaced by the second sound. Note that the value of  $x$ , where  $R(q, x)=0$  at 11°K, corresponds roughly to  $d_{II}=0.596$ , as it can be expected from (4.6) for  $\tau_R \gg \tau_N$ .

branches. Different purities have been simulated by varying a parameter  $A_I$  which determines part of the resistive scattering rate. We found the following results for the possible modes of heat transport through a dielectric crystal.

(a) For  $\bar{\Omega}\tau \gg 1$  heat is carried by single ballistic phonons. It is partly a semantic question whether this form of heat pulses is a collective or a single-

particle excitation. In the framework of this analysis the latter terminology seems to be more adequate, in contrast to Ranninger's opinion.<sup>20</sup>

(b) For  $\bar{\Omega}\tau_N \lesssim 1$  and  $\bar{\Omega}\tau_R \gtrsim 1$  heat can propagate as second sound. Its velocity  $C_2$  will be a decreasing function of temperature and finally go to zero as  $\tau_R\bar{\Omega} \lesssim 2$ . This observation is similar to Ranninger's result. Our variation of  $C_2$  seems, however, to be stronger.

(c) For given  $\bar{\Omega}$  there are critical values of  $\tau_N$  and  $\tau_R$  such that second sound can only propagate for  $QC_t\tau_N < (QC_t\tau_N)_c$  and  $QC_t\tau_R < (QC_t\tau_R)_c$ . Second sound is therefore a collective mode which is distinct from ballistic heat pulses. The velocity  $C_2$  at the point where second sound starts developing is the lower the higher the ratio  $(\tau_N)_c/(\tau_R)_c$ .

It is our hope that the present work has added some more realistic features to the previous treatments of the transition from the ballistic regime of heat conduction to second sound. We also suggest that the general results of Sec. III can be used to get more reliable information about the temperature dependence of the relaxation times involved, e.g., by comparing such quantities as  $C_2(T)$  with the experimental data. On the other hand, it is clear that some of the facts listed in Sec. V may be due to the approximations made in our calculation. A realistic anisotropic phonon spectrum would certainly modify the details, although it would hardly affect the general behavior. The most serious approximation consists, of course, in replacing the true integral operator  $C$  by two relaxation times. It is therefore planned to investigate analytically and numerically what results could be derived from the Boltzmann equation (2.6) using the true eigenfunctions of  $C$ .

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## Theory of Multiphonon Absorption due to Anharmonicity in Crystals

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A theory of multiphonon absorption due to anharmonicity in crystals is presented, employing a Green's-function technique. In contrast to previous theories, we do *not* expand the lattice-interaction potential in powers of displacements. We are therefore able to obtain a single expression for the absorption coefficient  $\alpha$ , which includes various classes of contributions to infinite order in phonons, and which is valid for all frequencies in the multiphonon regime. The results involve just the displacement-correlation tensor of the lattice, and the Fourier components of the interatomic potential  $v$ . Simplified expressions are obtained for isotropic models, and specific choices of  $v$ . Within the Einstein model, one finds an exponential behavior,  $\alpha(\omega) \sim e^{-\sigma\omega}$ ; predicted values of  $\sigma$  within the model are found to be in good agreement with recent experimental data on alkali halides. A general technique for evaluating  $\alpha$ , the method of convolutions, is elaborated. Predictions regarding the frequency and temperature dependence of  $\alpha$  are discussed and compared with other work.

### I. INTRODUCTION

Multiphonon absorption in crystals has its origin in two types of physical interactions: anharmonicity and higher-order electric moments.<sup>1</sup> While the electric-moment interaction allows for direct excitation of phonons by light, the anharmonic interaction must act in conjunction with the electric-moment interaction to produce absorption. A number of papers have calculated the absorption due to anharmonicity<sup>2</sup> and higher-order electric moments<sup>3</sup> separately, as well as in combination with each other.<sup>4</sup> From the results of these calculations it is not evident under just what conditions anharmonic effects, as opposed to those due to higher-order electric moments, dominate. In recent work on the latter question, it was found<sup>5</sup> that, for low-

order multiphonon processes, either mechanism may dominate within a range of actual representative materials.

There has been much interest recently in connection with highly transparent ultrapure infrared materials, which, for example, are necessary to provide relatively distortion-free optical elements for high-power laser applications.<sup>6</sup> In practice, the useful frequency range of such materials is the many-phonon regime above the fundamental phonon frequency, but well below the electronic gap. Past calculations<sup>1-5</sup> of multiphonon effects have been concerned chiefly with photon frequencies near resonance, and have employed perturbative expansions of the lattice potential in powers of lattice displacements in order to calculate the quantities of interest. Such methods become extremely