would then be the 6–12 nearest-neighbor-model. Horton and Leech[9] also concluded that near-neighbor calculations best fit the data for argon and krypton.

A nearest-neighbor model is physically unrealistic. It assumes that the potential vanishes abruptly at a distance quite close to the potential well. One would expect a meaningful theoretical model to incorporate all neighbor interactions. As pointed out by Guggenheim and McGlashan,[33] the Lennard-Jones potential exaggerates the effect of distant neighbors and is inadequate for predicting the thermal properties of the solids. What is required is a deeper and shorter range potential. In this connection, Leech and Reissland[34] have added an intermediate-range attractive term $1/r^8$ (i.e., the next term in the expansion of the Van der Waal's potential) to the Lennard-Jones 6-12 potential. The agreement of their specific-heat calculations with Morrison's argon data[5] was not significantly better than the Horton and Leech 6–12 (AN) results. However, a $1/r^9$ term[35] considerably improved the agreement with the argon experiments. The effect of this term is to deepen the potential well and hence decrease the effect of all but the nearest neighbors. It would be interesting to see how well theoretical calculations of the specific heat, with this new term in the potential, compare with our xenon and neon data.

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Unified Approach to Interacting Phonon Problems*

Philip C. Kwok† and Paul C. Martin

Lyman Laboratory of Physics, Harvard Univessity, Cambridge, Massachusetts

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A unified treatment of the behavior of a system of interacting phonons is presented. This treatment is based on the physically measurable atomic-displacement correlation function or structure function. The perturbation treatment of its high-frequency behavior is briefly summarized. Its more complex behavior at low frequencies is studied in detail. Using a simple model of longitudinal phonons with cubic anharmonic interactions and without umklapp processes, two modes, representing damped first and second sound, are obtained. The parameters which occur in the calculated correlation function are shown to agree with those expected more generally from a phenomenological analysis which is also presented. The paper clarifies certain paradoxes relating to the difference between phonon, ordinary sound, and second sound, by showing that there is no fundamental distinction between these concepts in the only physical quantity, the displacement correlation function.

I. INTRODUCTION

As is by now well known, inelastic neutron scattering and Brillouin scattering measure the distribution in energy of density fluctuations of an atomic system. At high frequencies when these fluctuations have a wavelength short compared to an average mean free path this continuous energy spectrum has a well-defined peak. The position and width of this peak correspond to the natural frequency and damping of the phonon or the ordinary sound mode. The collision rate $1/\tau$ which corresponds to this mean free path is approximately equal to the damping of thermal frequency phonons. When the fluctuations have a wavelength long compared with the mean free path or $\omega\tau \ll 1$, a hydrodynamic description applies. In this limit, there appears a second peak with a smaller weight. It describes the thermal conduction mode or under certain circum-

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† Present address: IBM Watson Research Center, Yorktown Heights, New York.

stances the second sound mode associated with propagation of phonon energy density. From a theoretical point of view the behavior of the high-frequency excitation spectrum is easily understood: The infinitely long-lived, "theoretical" phonon is simply the normal mode of the atomic vibration; its damping is determined by a standard quantum-mechanical perturbation calculation of the anharmonic interactions between these "theoretical" phonons.[1] On the other hand, in the hydrodynamic limit, the problem is treated in a different and incomplete fashion. One introduces without microscopic derivation the ordinary sound mode with velocity determined by the *adiabatic* elastic constants. One calculates the attenuation of this wave due to the irreversible processes it produces in a phonon heat bath from a phenomenological Boltzmann equation for the phonon distribution function.[2] The second sound mode is derived from the conservation laws implied by this equation.[3] To find the relationship between these two hydrodynamic modes, i.e., their relative weights in the density fluctuation excitation spectrum, one has to make use of another theoretical construct, namely, the two-fluid hydrodynamics.[4]

The hydrodynamic region is a complicated one if its description is based on the "theoretical" or harmonic phonon because the hydroydanmic modes involve the interactions of many such phonons. On the other hand, from the point of view of the experimentalist, who sees "physical" phonons, no such difficulty arises. The purpose of this paper is to present a unified "physical" treatment which eliminates the deficiencies of the usual hydrodynamic treatment and goes over to the familiar treatment at high frequencies where the "theoretical" phonon and "physical" phonon are the same. The "physical" phonon is described by the displacement correlation function or phonon Green's function which we calculate by using thermodynamic Green's-function techniques. We show, in particular, that this function has two oscillation frequencies for long wavelengths when umklapp processes are unimportant, corresponding to the (damped) first and second sound. Since the natural frequencies of the displacement correlations are the phonons, our conclusion is tantamount to the statement that both first and second sound are phonons.

The calculations are carried out for a particularly simple model of the interacting phonons to avoid unnecessary complications. Furthermore, in this paper, we only consider the low-frequency limit, since the high-frequency calculations are more straightforward.

As an introduction let us review the conventional but nevertheless instructive hydrodynamic treatment based on the phenomenological Boltzmann equation

$$(\partial/\partial t)N(\mathbf{p}\mathbf{r}t)+c\mathbf{p}\cdot\boldsymbol{\nabla}_{\mathbf{r}}N(\mathbf{p}\mathbf{r}t)=-(\partial N/\partial t)_{\text{collision}}, \quad (1.1)$$

for the phonon distribution function $N(\mathbf{p}\mathbf{r}t)$ and trace the essential steps of the derivation of second sound. We begin with the conservation law for the heat density or energy density of the phonons q,

$$\partial q/\partial t+\boldsymbol{\nabla}\cdot\mathbf{j}_q=0, \quad (1.2)$$

where $\mathbf{j}_q$ is the heat current. If the heat current $\mathbf{j}_q$ is also a conserved quantity we have

$$(\partial/\partial t)j_{qi}+\nabla_j X_{ij}=0, \quad (1.3)$$

where X_{ij} is the heat flux tensor. At a frequency low enough that local equilibrium can be established it is possible to express X_{ij} entirely in terms of conserved quantities. Then in an isotropic medium where $X_{ij}=\delta_{ij}X$, we can combine (1.2) and (1.3) and obtain the following wave equation for q:

$$(\partial^2/\partial t^2)q-(\partial X/\partial q)_{\text{eq}}\nabla^2 q=0; \quad (1.4)$$

the velocity of the second sound u_2 is given by $(\partial X/\partial q)^{1/2}$ at equilibrium.

In a superfluid (e.g., liquid He^4) in the absence of vorticity the second sound mode exists because (1.2) and (1.3) are satisfied, the latter as a result of $\mathbf{j}_q$ being proportional to $\mathbf{v}_s$, the conserved superfluid velocity, in a reference frame in which the particle current is zero.[5] For the phonons in a crystal we also have second sound. The reason is that if we neglect the small anharmonic corrections to the expression for $\mathbf{j}_q$, then $\mathbf{j}_q$ is equal to $c^2\mathbf{P}$, where c is the sound velocity and $\mathbf{P}$ is the phonon momentum density, so that it also satisfies a conservation law. Specifically, we have

$$(\partial/\partial t)j_{qi}+\nabla_j c^2\Lambda_{ij}=-(1/\tau_U)j_{qi}, \quad (1.3')$$

where Λ_{ij} is the momentum stress tensor and τ_U is the collision time for the umklapp processes which do not conserve $\mathbf{P}$. The collision time gives rise to a finite thermal conductivity that damps the second sound mode. Other dissipative coefficients are obtained using the Boltzmann equation (1.1) and expanding $c^2\Lambda_{ij}=X_{ij}$ about its local equilibrium value $(X_{ij})_0$. These terms are schematically represented by

$$X_{ij}\cong(X_{ij})_0+D\delta_{ij}(\partial q/\partial t), \qquad D\cong c^2\tau. \quad (1.5)$$

Then from Eqs. (1.2), (1.3'), and (1.5) we obtain

$$\frac{\partial^2}{\partial t^2}q-\left(\frac{\partial X}{\partial q}\right)_{\text{eq}}\nabla^2 q-D\frac{\partial}{\partial t}\nabla^2 q+\frac{1}{\tau_U}\frac{\partial q}{\partial t}=0 \quad (1.6)$$

and, consequently, the following dispersion relation for

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[2] A. Akhiezer, J. Phys. (USSR) **1**, 277 (1939); see also T. O. Woodruff and H. Ehrenreich, Phys. Rev. **123**, 1553 (1961).
[3] J. C. Ward and J. Wilks, Phil. Mag. 43, 48 (1952).
[4] P. C. Hohenberg and P. C. Martin, Ann. Phys. (N. Y.) (to be published).

[5] The velocity of the normal fluid $\mathbf{v}_n$ is equal to $-(\rho_s/\rho_n)\mathbf{v}_s$ since $\rho_n\mathbf{v}_n+\rho_s\mathbf{v}_s=0$.

the propagation of heat waves

$$\omega^2 - u_2^2k^2 + i\omega(Dk^2 + 1/\tau_U) = 0, \quad (1.7)$$

$$u_2^2 = (\partial X/\partial q)_{\rm eq} \cong \tfrac{1}{3}c^2. \quad (1.8)$$

At very low frequencies where $\omega\tau_U \ll 1$, Eq. (1.7) describes thermal conduction as it is adequately approximated by $-u_2^2k^2 + i\omega/\tau_U = 0$. In the frequency region $\omega D \ll c^2$ and $\omega\tau_U \gg 1$, it yields the slightly damped second sound mode. In the subsequent analysis, Eq. (1.7) and the corresponding equation for first sound will appear as the natural frequencies of the displacement correlations.

In Sec. 2, the model is formulated and the Green's-function formulation briefly described. In Sec. 3, a generalized Boltzmann equation is derived for the nonequilibrium Green's function. In Sec. 4, the various equilibrium correlation functions are deduced directly from the Boltzmann equation and the equation for the nonequilibrium displacement. In Sec. 5, the displacement correlation function is rederived with a slight rearrangement to conform more closely to the derivation at high frequencies. The high-frequency results, and the interpolation between them and the results contained in this paper, have been summarized elsewhere[6] and are not reported here.

II. MATHEMATICAL FORMULATION OF THE MODEL

We will now study the various problems of the interacting phonons posed above by using a simple model with longitudinal phonons only. The Hamiltonian density for this model is

$$\begin{aligned}\mathcal{H}(\mathbf{r}t) = &\tfrac{1}{2}\pi^2(\mathbf{r}t) + \tfrac{1}{2}c^2\nabla d(\mathbf{r}t)\cdot\nabla d(\mathbf{r}t)\\ &-\tfrac{1}{4}\alpha(\pi(\mathbf{r}t)\nabla d(\mathbf{r}t)\cdot\nabla d(\mathbf{r}t) + \nabla d(\mathbf{r}t)\cdot\nabla d(\mathbf{r}t)\pi(\mathbf{r}t))\\ &+\tfrac{1}{8}\alpha^2(\nabla d(\mathbf{r}t)\cdot\nabla d(\mathbf{r}t)) - J(\mathbf{r}t)d(\mathbf{r}t), \quad (2.1)\end{aligned}$$

where $d(\mathbf{r}t)$ is the displacement field and π is the canonical conjugate field which satisfies the following commutation relation[7] with d

$$[\pi(\mathbf{r}t), d(\mathbf{r}'t)] = (1/i)\delta^3(\mathbf{r}-\mathbf{r}'). \quad (2.2)$$

α is an effective cubic anharmonic coupling constant and J is an external source introduced to generate the necessary equations. From (2.1) and (2.2) we readily find that

$$\dot{d}(\mathbf{r}t) = \pi(\mathbf{r}t) - \tfrac{1}{2}\alpha\nabla d(\mathbf{r}t)\cdot\nabla d(\mathbf{r}t), \quad (2.3)$$

and

$$\begin{aligned}\ddot{d}(\mathbf{r}t) = &c^2\nabla^2 d(\mathbf{r}t) - \alpha(\nabla\dot{d}(\mathbf{r}t)\cdot\nabla d(\mathbf{r}t) + \nabla d(\mathbf{r}t)\cdot\nabla\dot{d}(\mathbf{r}t)\\ &+\tfrac{1}{2}\dot{d}(\mathbf{r}t)\nabla^2 d(\mathbf{r}t) + \tfrac{1}{2}\nabla^2 d(\mathbf{r}t)\dot{d}(\mathbf{r}t)) + J(\mathbf{r}t). \quad (2.4)\end{aligned}$$

Following the conventional procedure,[8] we define the ensemble average of any operator $A(\mathbf{r}t)$ (with t in the imaginary time interval $\{0, -i\beta\}$, $\beta = 1/k_BT$) in the presence of J as

$$\langle A(\mathbf{r}t)\rangle = \frac{\mathrm{Trace}\, e^{-\beta H_0}(SA(\mathbf{r}t))_+}{\mathrm{Trace}\, e^{-\beta H_0}S}, \quad (2.5)$$

where $H_0 = \int d^3r\, \mathcal{H}(\mathbf{r}t)$ with $J=0$, and S is the time-ordered operator

$$S = \left(\exp i\int_0^{\tau_0} dt \int d^3r\, J(\mathbf{r}t)d(\mathbf{r}t)\right)_+, \quad (2.6)$$

$$\tau_0 = -i\beta.$$

The desired physical functions of real time are obtained by a trivial analytic continuation. The Green's function for the displacement field is defined as

$$G(\mathbf{r}t;\mathbf{r}'t') = i(\langle(d(\mathbf{r}t)d(\mathbf{r}'t'))_+\rangle - \langle d(\mathbf{r}t)\rangle\langle dZ\mathbf{r}'t'\rangle) \quad (2.7)$$

or, equivalently, using (2.5) and (2.6), as

$$G(\mathbf{r}t;\mathbf{r}'t') = \frac{\delta\langle d(\mathbf{r}t)\rangle}{\delta J(\mathbf{r}'t')} = \frac{\delta\langle d(\mathbf{r}'t')\rangle}{\delta J(\mathbf{r}t)}. \quad (2.8)$$

We obtain the equation for G by first taking the ensemble average of (2.4), getting

$$\begin{aligned}&(\partial^2/\partial t_1^2)\langle d(1)\rangle - c^2\nabla_1^2\langle d(1)\rangle\\ &\quad + (1/2i)\alpha(123)(G(23) + \langle d(2)\rangle\langle d(3)\rangle) = J(1). \quad (2.9)\end{aligned}$$

In this equation, we have introduced

$$\begin{aligned}\alpha(123) = -2\alpha\bigg(&\frac{\partial}{\partial t_2}\nabla_2\delta(12)\cdot\nabla_3\delta(13)\\ &+\nabla_2\delta(12)\cdot\nabla_3\frac{\partial}{\partial t_3}\delta(13) + \frac{1}{2}\frac{\partial}{\partial t_2}\delta(12)\nabla_3^2\delta(13)\\ &+\tfrac{1}{2}\nabla_2^2\delta(12)\frac{\partial}{\partial t_3}\delta(13)\bigg) \quad (2.10)\end{aligned}$$

and also have used the abbreviation $\{1\} \leftrightarrow \{\mathbf{r}_1, t_1\}$, etc., and the repeated index summation convention; we then differentiate (2.9) with respect to J:

$$((\partial^2/\partial t_1^2) - c^2\nabla_1^2)G(11') - \Sigma(12)G(21') = \delta(11'). \quad (2.11)$$

Σ is the self-energy operator that contains all the anharmonic corrections

$$\Sigma(11') = -\alpha(11'2)\langle d(2)\rangle - \frac{1}{2i}\alpha(123)\frac{\delta G(23)}{\delta\langle d(1')\rangle}. \quad (2.12)$$

This equation can be written in the following iterative form:

$$\begin{aligned}\Sigma(11') = &-\alpha(11'2)\langle d(2)\rangle\\ &-\frac{1}{2i}\alpha(123)G(2\bar{2})G(3\bar{3})\left(\frac{\delta\Sigma(\bar{2}\bar{3})}{\delta\langle d(1')\rangle}\right), \quad (2.13)\end{aligned}$$

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[6] P. C. Kwok, P. C. Martin, and P. B. Miller, Solid State Commun. (to be published); P. C. Kwok, thesis, Harvard University, 1965 (unpublished).

[7] We take $\hbar = 1$.

[8] See, for example, L. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (W. A. Benjamin, Inc., New York, 1962).

with the help of the identities

$$\frac{\delta G(23)}{\delta\langle d(1')\rangle}=-G(2\bar{2})\frac{\delta G^{-1}(\bar{2}\bar{3})}{\delta\langle d(1')\rangle}G(\bar{3}3)\,,$$
$$G^{-1}(11')=((\partial^2/\partial t_1{}^2)-c^2\nabla_1{}^2)\delta(11')-\Sigma(11')\,. \quad (2.14)$$

The perturbation series for Σ can now be easily obtained from (2.13) by carrying out the indicated functional derivative. After this is done we can put the source $J=0$. Then $\langle d\rangle$ vanishes, and in the ensuing spatially invariant system $G(11')$ and $\Sigma(11')$ depend only on t_1-t_1' and $|\mathbf{r}_1-\mathbf{r}_1|$.

In this paper, we will not pursue the perturbative treatment, since it only involves calculations of successive collisions. Our primary aim is to study the collision-dominated or hydrodynamic region, obtaining the second sound mode, and to examine the various low-frequency properties connected with it. For this purpose, it is necessary to deduce a Boltzmann-like equation.

III. GENERALIZED BOLTZMANN EQUATION

As discussed by Kadanoff and Baym,[8] the derivation of the Boltzmann equation is best carried out directly in terms of the real time Green's function $G^>$ or $G^<$ defined as

$$G^>(11')=i(\langle d(1)d(1')\rangle-\langle d(1)\rangle\langle d(1')\rangle)\,,$$
$$G^<(11')=G^>(1'1)\,. \quad (3.1)$$

In thermal equilibrium, the system is uniform and the functions $G^{>,<}(11')$ depend only on the relative coordinates $\{1-1'\}$. We can then define the following Fourier transform:

$$G^{>,<}(11')=i\int\frac{d\omega}{2\pi}\int\frac{d^3p}{(2\pi)^3}e^{i\mathbf{p}\cdot(\mathbf{r}_1-\mathbf{r}_1')-i\omega(t_1-t_1')}\times G^{>,<}(\mathbf{p},\omega)\,. \quad (3.2)$$

The transforms $G^{>,<}(\mathbf{p}\omega)$ can be readily shown to have the following representation[8]:

$$G^>(\mathbf{p}\omega)=2\chi''(\mathbf{p}\omega)[1+N_0(\omega)]\,,$$
$$G^<(\mathbf{p}\omega)=2\chi''(\mathbf{p}\omega)N_0(\omega)\,, \quad (3.3)$$

where $N_0(\omega)$ is the equilibrium phonon distribution function

$$N_0(\omega)=(e^{\beta\omega}-1)^{-1}\,, \quad (3.4)$$

and $\chi''(\mathbf{p}\omega)$ is the spectral function giving the frequency distribution of the elementary excitation of the system. In the harmonic approximation in which $\alpha=0$, χ'' is equal to

$$\chi_0''(\mathbf{p}\omega)=\pi(\omega/|\omega|)\delta(\omega^2-c^2p^2)\,. \quad (3.5)$$

Let us now consider the system not in complete thermal equilibrium. The system will no longer be uniform. Therefore $G^{>,<}(11')$ are not just functions of $\{1-1'\}$, but depend on both 1 and $1'$. If we describe these functions by the relative coordinates $\mathbf{r}=\mathbf{r}_1-\mathbf{r}_1'$, $t=t_1-t_1'$, and the center-of-mass coordinates $\mathbf{R}=\frac{1}{2}(\mathbf{r}_1+\mathbf{r}_1')$, $\hat{T}=\frac{1}{2}(t_1+t_1')$, we can still use the definition (3.2). However, the Fourier transforms $G^{>,<}(\mathbf{p}\omega)$ must now be dependent on $\mathbf{R}$ and $\hat{T}$, so that

$$G^{>,<}(11')=i\int\frac{d\omega}{2\pi}\int\frac{d^3p}{(2\pi)^3}e^{i\mathbf{p}\cdot\mathbf{r}-i\omega t}G^{>,<}(\mathbf{p}\omega,\mathbf{R}\hat{T})\,. \quad (3.2')$$

Likewise, Eq. (3.3) becomes

$$G^>(\mathbf{p}\omega,\mathbf{R}\hat{T})=2\chi''(\mathbf{p}\omega,\mathbf{R}\hat{T})[1+N(\mathbf{p}\omega,\mathbf{R}\hat{T})]\,,$$
$$G^<(\mathbf{p}\omega,\mathbf{R}\hat{T})=2\chi''(\mathbf{p}\omega,\mathbf{R}\hat{T})N(\mathbf{p}\omega,\mathbf{R}\hat{T})\,. \quad (3.3')$$

The functions $\chi''(\mathbf{p}\omega,\mathbf{R}\hat{T})$ and $N(\mathbf{p}\omega,\mathbf{R}\hat{T})$ are easily interpreted as the local spectral function and the local phonon distribution function, respectively. To find the equation for $G^>(\mathbf{p}\omega,\mathbf{R}\hat{T})$ or $G^<(\mathbf{p}\omega,\mathbf{R}\hat{T})$, we begin with Eq. (2.11) and a similar equation with 1 and $1'$ interchanged, namely,

$$((\partial^2/\partial t_{1'}{}^2)-c^2\nabla_{1'}{}^2)G(11')-\Sigma(1'2)G(21)=\delta(11')\,. \quad (3.6)$$

Subtracting (3.6) from (2.11), we obtain

$$\left[\left(\frac{\partial}{\partial t_1}+\frac{\partial}{\partial t_{1'}}\right)\left(\frac{\partial}{\partial t_1}-\frac{\partial}{\partial t_{1'}}\right)-c^2(\nabla_1+\nabla_{1'})\cdot(\nabla_1-\nabla_{1'})\right]G(11')$$
$$+\alpha(132)\langle d(3)\rangle G(21')-\alpha(1'32)\langle d(3)\rangle G(21)=\Sigma'(12)G(21')-\Sigma'(1'2)G(21)\,, \quad (3.7)$$

where $\Sigma'(12)$ is the self-energy operator minus the "source" term $\{-\alpha(123)\langle d(3)\rangle\}$. Our next step is to change the coordinates 1 and $1'$ to $\mathbf{r}t$ and $\mathbf{R}\hat{T}$ and take the Fourier transform with respect to $\mathbf{r}$ and t. The resulting equation is very complicated, unless the deviation from thermal equilibrium is small and slowly varying in space and time. In that case, it can be simplified by expanding the various terms in powers of the gradient $\nabla_\mathbf{R}$ and time derivative $\partial/\partial\hat{T}$. Then, keeping only the lowest order terms, we find

$$\left(\frac{\partial}{\partial\hat{T}}+\nabla_\mathbf{R}\cdot c^2\mathbf{p}\right)G^{>,<}(\mathbf{p}\omega,\mathbf{R}\hat{T})-\alpha\left[(\nabla_{R_i}\nabla_{R_j}\langle d(\mathbf{R}\hat{T})\rangle)\omega p_i\frac{\partial}{\partial p_j}-\frac{1}{2}\left(\nabla_{R_i}\frac{\partial}{\partial\hat{T}}\langle d(\mathbf{R}\hat{T})\rangle\right)p^2\frac{\partial}{\partial p_i}\right]G^{>,<}(\mathbf{p}\omega,\mathbf{R}\hat{T})$$
$$+\alpha\left[\left(\nabla_{R_i}\frac{\partial}{\partial\hat{T}}\langle d(\mathbf{R}\hat{T})\rangle\right)p_i\omega\frac{\partial}{\partial\omega}-\frac{1}{2}\left(\frac{\partial^2}{\partial\hat{T}^2}\langle d(\mathbf{R}\hat{T})\rangle\right)p^2\frac{\partial}{\partial\omega}\right]G^{>,<}(\mathbf{p}\omega,\mathbf{R}\hat{T})$$
$$=-\tfrac{1}{2}(\Sigma'^>(\mathbf{p}\omega,\mathbf{R}\hat{T})G^<(\mathbf{p}\omega,\mathbf{R}\hat{T})-\Sigma'^<(\mathbf{p}\omega,\mathbf{R}\hat{T})G^>(\mathbf{p}\omega,\mathbf{R}\hat{T}))\,, \quad (3.8)$$

where $\Sigma'^{>,<}$ are functions similar to the $G^{>,<}$ defined from Σ'.

First of all, we examine the equation satisfied by the local spectral function $\chi''(\mathbf{p}\omega,\mathbf{R}\hat{T})$ that can be readily obtained from (3.8) with the help of (3.3′):

$$\left[\left(\frac{\partial}{\partial\hat{T}}\omega+\nabla_{\mathbf{R}}\cdot c^2\mathbf{p}\right)-\alpha(\nabla_i\nabla_j\langle d\rangle)\omega p_i\frac{\partial}{\partial p_j}+\tfrac{1}{2}\alpha\left(\nabla_i\frac{\partial}{\partial\hat{T}}\langle d\rangle\right)p^2\frac{\partial}{\partial p_i}\right.$$
$$\left.+\alpha\left(\nabla_i\frac{\partial}{\partial\hat{T}}\langle d\rangle\right)p_i\omega\frac{\partial}{\partial\omega}-\tfrac{1}{2}\alpha\left(\frac{\partial^2}{\partial\hat{T}^2}\langle d\rangle\right)p^2\frac{\partial}{\partial\omega}\right]\chi''(\mathbf{p}\omega,\mathbf{R}\hat{T})=0. \quad (3.9)$$

Clearly, to first order in $\langle d\rangle$,

$$\chi''(\mathbf{p}\omega,\mathbf{R}\hat{T})=f\left[(\omega-\alpha\mathbf{p}\cdot\nabla_{\mathbf{R}}\langle d(\mathbf{R}\hat{T})\rangle)^2-\left(c-\frac{\alpha}{2c}\frac{\partial}{\partial\hat{T}}\langle d(\mathbf{R}\hat{T})\rangle\right)^2p^2\right]; \quad (3.10)$$

for any arbitrary function f is a solution to (3.9). To determine f, we use the fact that when $\langle d\rangle=0$ it must reduce to the thermal equilibrium spectral function, which is simply χ_0'' in our present approximation because we have neglected in (3.8) terms that give rise to anharmonic corrections to the spectral function.[9] Thus we find that

$$\chi''(\mathbf{p}\omega,\mathbf{R}\hat{T})=\pi\frac{\omega-\alpha\mathbf{p}\cdot\nabla_{\mathbf{R}}\langle d\rangle}{|\omega-\alpha\mathbf{p}\cdot\nabla_{\mathbf{R}}\langle d\rangle|}\delta\left[(\omega-\alpha\mathbf{p}\cdot\nabla_{\mathbf{R}}\langle d\rangle)^2-\left(c-\frac{\alpha}{2c}\langle\dot{d}\rangle\right)^2p^2\right]. \quad (3.11)$$

If we now go back to Eq. (3.8) and observe that we can extract the spectral function χ'' from both sides, we immediately obtain the following equation for the phonon distribution function $N(\mathbf{p}\omega,\mathbf{R}\hat{T})$, which is the Boltzmann-like equation we seek:

$$\left(\frac{\partial}{\partial\hat{T}}\omega+\nabla_{\mathbf{R}}\cdot c^2\mathbf{p}\right)N(\mathbf{p}\omega,\mathbf{R}\hat{T})-\alpha\left[(\nabla_{R_i}\nabla_{R_j}\langle d\rangle)\omega p_i\frac{\partial}{\partial p_j}-\frac{1}{2}\left(\nabla_{R_i}\frac{\partial}{\partial\hat{T}}\langle d\rangle\right)p^2\frac{\partial}{\partial p_i}\right]N(\mathbf{p}\omega,\mathbf{R}\hat{T})+\alpha\left[\left(\nabla_{R_i}\frac{\partial}{\partial\hat{T}}\langle d\rangle\right)p_i\omega\frac{\partial}{\partial\omega}\right.$$
$$\left.-\frac{1}{2}\left(\frac{\partial^2}{\partial\hat{T}^2}\langle d\rangle\right)p^2\frac{\partial}{\partial\omega}\right]N(\mathbf{p}\omega,\mathbf{R}\hat{T})=-\tfrac{1}{2}[\Sigma'^{>}(\mathbf{p}\omega,\mathbf{R}\hat{T})N(\mathbf{p}\omega,\mathbf{R}\hat{T})-\Sigma'^{<}(\mathbf{p}\omega,\mathbf{R}\hat{T})(1+N(\mathbf{p}\omega,\mathbf{R}\hat{T}))]. \quad (3.12)$$

The description will be complete when $\Sigma'^{>}$ and $\Sigma'^{<}$ are known. These functions can be easily calculated, if we retain only the lowest order term in the perturbation series for Σ', which is, according to (2.13),

$$\Sigma'(11')=(1/2i)\alpha(123)G(2\bar{2})G(3\bar{3})\alpha(\bar{2}\bar{3}1'). \quad (3.13)$$

They are

$$\Sigma'^{>,<}(\mathbf{p}\omega,\mathbf{R}\hat{T})=\alpha^2\int\frac{d\omega_1}{2\pi}\int\frac{d^3p_1}{(2\pi)^3}\int\frac{d\omega_2}{2\pi}\int\frac{d^3p_2}{(2\pi)^3}(2\pi)^4\delta(\omega_1-\omega_2-\omega)\delta^3(\mathbf{p}_1-\mathbf{p}_2-\mathbf{p})$$
$$\times[(\omega_1-\omega_2)\mathbf{p}_1\cdot\mathbf{p}_2-\tfrac{1}{2}\omega_1p_2^2+\tfrac{1}{2}\omega_2p_1^2]^2G^{>,<}(\mathbf{p}_1\omega_1,\mathbf{R}\hat{T})G^{>,<}(\mathbf{p}_2\omega_2,\mathbf{R}\hat{T}). \quad (3.14)$$

Substituting them into (3.12), we finally obtain

$$\left[\left(\frac{\partial}{\partial\hat{T}}\omega+\nabla_{\mathbf{R}}\cdot c^2\mathbf{p}\right)-\alpha(\nabla_i\nabla_j\langle d\rangle)\omega p_i\frac{\partial}{\partial p_j}+\tfrac{1}{2}\alpha\left(\nabla_i\frac{\partial}{\partial\hat{T}}\langle d\rangle\right)p^2\frac{\partial}{\partial p_i}+\alpha\left(\nabla_i\frac{\partial}{\partial\hat{T}}\langle d\rangle\right)p_i\omega\frac{\partial}{\partial\omega}-\tfrac{1}{2}\alpha\left(\frac{\partial^2}{\partial\hat{T}^2}\langle d\rangle\right)p^2\frac{\partial}{\partial\omega}\right)\right]N(\mathbf{p}\omega,\mathbf{R}\hat{T})$$
$$=-\sigma(\mathbf{p}\omega,\mathbf{R}\hat{T})$$
$$=-\alpha^2\int\frac{d\omega_1}{2\pi}\int\frac{d^3p_1}{(2\pi)^3}\int\frac{d\omega_2}{2\pi}\int\frac{d^3p_2}{(2\pi)^3}(2\pi)^4\delta(\omega_1-\omega_2-\omega)\delta^3(\mathbf{p}_1-\mathbf{p}_2-\mathbf{p})$$
$$\times[(\omega_1-\omega_2)\mathbf{p}_1\cdot\mathbf{p}_2-\tfrac{1}{2}\omega_1p_2^2+\tfrac{1}{2}\omega_2p_1^2]^2 2\chi''(\mathbf{p}_1\omega_1,\mathbf{R}\hat{T})2\chi''(\mathbf{p}_2\omega_2,\mathbf{R}\hat{T})[(1+N(\mathbf{p}_1\omega_1,\mathbf{R}\hat{T}))N(\mathbf{p}_2\omega_2,\mathbf{R}\hat{T})N(\mathbf{p}\omega,\mathbf{R}\hat{T})$$
$$-N(\mathbf{p}_1\omega_1,\mathbf{R}\hat{T})(1+N(\mathbf{p}_2\omega_2,\mathbf{R}\hat{T}))(1+N(\mathbf{p}\omega,\mathbf{R}\hat{T}))]. \quad (3.15)$$

We will now study the energy and momentum conservation laws. The energy and momentum density can be

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[9] These are the "Poisson-bracket" terms in the better generalized Boltzmann equation described in Ref. 8, Chap. 9.

readily found from the Hamiltonian (2.1) to be

$$E(\mathbf{R}\hat{T})=\int\frac{d\omega}{2\pi}\int\frac{d^3p}{(2\pi)^3}\tfrac{1}{2}(\omega^2+c^2p^2)G^<(\mathbf{p}\omega,\mathbf{R}\hat{T}) \tag{3.16}$$

and

$$\mathbf{P}(\mathbf{R}\hat{T})=\int\frac{d\omega}{2\pi}\int\frac{d^3p}{(2\pi)^3}\omega\mathbf{p}G^<(\mathbf{p}\omega,\mathbf{R}\hat{T}). \tag{3.17}$$

Working with these expressions for the energy and momentum is inconvenient since they contain parts that depend explicitly on the displacement of the atoms making up the crystal. Since we will only study linear effects, we can separate these parts out. For example, the energy density (3.16) can be written in the following way:

$$E(\mathbf{R}\hat{T})=\int\frac{d\omega}{2\pi}\int\frac{d^3p}{(2\pi)^3}\tfrac{1}{2}(\omega^2+c^2p^2)2(\chi''(\mathbf{p}\omega,\mathbf{R}\hat{T})-\chi_0''(\mathbf{p}\omega))(N(\mathbf{p}\omega,\mathbf{R}\hat{T})-N_0(\omega))$$
$$+\int\frac{d\omega}{2\pi}\int\frac{d^3p}{(2\pi)^3}\tfrac{1}{2}(\omega^2+c^2p^2)2(\chi''(\mathbf{p}\omega,\mathbf{R}\hat{T})-\chi_0''(\mathbf{p}\omega))N_0(\omega)+\int\frac{d\omega}{2\pi}\int\frac{d^3p}{(2\pi)^3}\tfrac{1}{2}(\omega^2+c^2p^2)2\chi_0''(\mathbf{p}\omega)N(\mathbf{p}\omega,\mathbf{R}\hat{T}). \tag{3.18}$$

Clearly, the first term can be neglected as it is of second order. The second term is the one that depends explicitly on $\langle d\rangle$, while the last term is the local thermal phonon energy. A similar separation can be carried out for $P_i(\mathbf{R}T)$. Therefore, denoting $\{P_i(\mathbf{R}\hat{T}),E(\mathbf{R}\hat{T})\}$ as $P_\mu(\mathbf{R}\hat{T})$, we have

$$P_\mu(\mathbf{R}\hat{T})=P_\mu^{(1)}(\mathbf{R}\hat{T})+P_\mu^{(2)}(\mathbf{R}\hat{T}),$$

where

$$P_\mu^{(1)}(\mathbf{R}\hat{T})=\int\frac{d\omega}{2\pi}\int\frac{d^3p}{(2\pi)^3}\omega p_\mu 2(\chi''(\mathbf{p}\omega,\mathbf{R}\hat{T})-\chi_0''(\mathbf{p}\omega))N_0(\omega),$$
$$P_\mu^{(2)}(\mathbf{R}\hat{T})=\int\frac{d\omega}{2\pi}\int\frac{d^3p}{(2\pi)^3}\omega p_\mu\chi_0''(\mathbf{p}\omega)N(\mathbf{p}\omega,\mathbf{R}\hat{T}), \tag{3.19}$$

and $p_\mu=\{p_i,[(\omega^2+c^2p^2/2\omega]\}$.

The $P_\mu^{(1)}$ can be calculated directly if χ'' is known. In particular, using the approximate form (3.11),[10] we find

$$E_1(\mathbf{R}\hat{T})=\frac{\alpha}{2c^2}\left(\frac{\partial}{\partial\hat{T}}\langle d(\mathbf{R}\hat{T})\rangle\right)(\epsilon_0+\epsilon_{T_0}),$$
$$\mathbf{P}_1(\mathbf{R}\hat{T})=\frac{\alpha}{c^2}(\nabla_\mathbf{R}\langle d(\mathbf{R}\hat{T})\rangle)(\tfrac{1}{3}\epsilon_0-\epsilon_{T_0}), \tag{3.20}$$

where $(\epsilon_0+\epsilon_{T_0})$ is the thermal equilibrium energy density of the phonons at temperature T_0 and ϵ_0 is the zero-point energy. To determine the $P_\mu^{(2)}$, we multiply Eq. (3.15) by p_μ and integrate over ω and p. Neglecting terms proportional to $\langle d\rangle^2$ and terms proportional to α^3 which come from expanding χ'' about χ_0'' in σ, we obtain

$$\frac{\partial E_2(\mathbf{R}\hat{T})}{\partial\hat{T}}+\nabla_\mathbf{R}\cdot\mathbf{j}_{E_2}=-\frac{2\alpha}{c^2}\left(\frac{\partial^2}{\partial\hat{T}^2}\langle d(\mathbf{R}\hat{T})\rangle\right)\epsilon_{T_0} \tag{3.21}$$

and

$$\frac{\partial P_{2i}(\mathbf{R}\hat{T})}{\partial\hat{T}}+\nabla_{R_j}\Lambda_{2ij}(\mathbf{R}\hat{T})=\frac{4}{3}\frac{\alpha}{c^2}\left(\nabla_{R_i}\frac{\partial}{\partial\hat{T}}\langle d(R\hat{T})\rangle\right)\epsilon_{T_0}, \tag{3.22}$$

with

$$\mathbf{j}_{E_2}(\mathbf{R}\hat{T})=c^2\mathbf{P}_2(\mathbf{R}\hat{T}),$$
$$\Lambda_{2ij}(\mathbf{R}\hat{T})=\int\frac{d\omega}{2\pi}\int\frac{d^3p}{(2\pi)^3}p_ip_j\times 2\chi_0''(\mathbf{p}\omega)N(\mathbf{p}\omega,\mathbf{R}\hat{T}). \tag{3.23}$$

The energy current therefore satisfies a conservation law. According to the general discussion in the Introduction there will be a second sound mode. We can see this more explicitly by carrying out the expansion of the various functions about their local equilibrium values. We begin by separating the distribution function

[10] These results for E_1 and $\mathbf{P}_1$ can only serve to indicate qualitatively their dependence on $\langle d\rangle$. Our approximate Boltzmann equation (3.8) and spectral function (3.11) are not accurate enough for these functions to agree quantitatively with conservation laws for E_1 and P_1 which can be derived from subtracting the equations (3.21) and (3.22) from a similar set for the total $E(\mathbf{R}t)$ and $\mathbf{P}(\mathbf{R}t)$. To remedy the situation, more terms in the Boltzmann equation must be included (for example, the "Poisson bracket" terms) so that a better spatial function together with better equations for E and $\mathbf{P}$ can be obtained. However, since the second sound mode in the various correlation functions occurs through E_2 and $\mathbf{P}_2$, the more complicated treatment is not necessary for our purpose.

$N(\mathbf{p}\omega,\mathbf{R}\hat{T})$ into two parts

$$N(\mathbf{p}\omega,\mathbf{R}\hat{T})=N_{\mathrm{QE}}(\mathbf{p}\omega,\mathbf{R}\hat{T})+N_1(\mathbf{p}\omega,\mathbf{R}\hat{T})\,,\quad(3.24)$$

where N_{QE} is the local- or quasi-equilibrium distribution function that makes the collision term σ in the Boltzmann equation vanish. From (3.15), we find that N_{QE} has the following form:

$$N_{\mathrm{QE}}(\mathbf{p}\omega,\mathbf{R}\hat{T})=[\exp(\beta(\mathbf{R}\hat{T})\omega-\beta_0\mathbf{P}\cdot\mathbf{V}(\mathbf{R}\hat{T}))-1]^{-1},\quad(3.25)$$

where $\beta_0=1/k_BT_0$. Here $\beta(\mathbf{R}\hat{T})$ and $\mathbf{V}(\mathbf{R}\hat{T})$ can be interpreted as the local temperature and the phonon drift velocity if $|\beta(\mathbf{R}\hat{T})-\beta_0|\ll\beta_0$ and $|\mathbf{V}(\mathbf{R}\hat{T})|\ll$ the sound velocity c. These parameters are not independent of each other, but are related through the conservation equations (3.21) and (3.22), in which we approximate the functions by their quasi-equilibrium values. Our next step is to calculate $N_1(\mathbf{p}\omega,\mathbf{R}\hat{T})$ by successive approximation, making use of the Boltzmann equation (3.8). The analysis is straightforward, and we obtain to linear order in the collision time τ_{ph}

$$E_2(\mathbf{R}\hat{T})=E_{2\mathrm{QE}}(\mathbf{R}\hat{T})\,,$$
$$\mathbf{j}_{E_2}(\mathbf{R}\hat{T})=c^2\mathbf{P}_2(\mathbf{R}\hat{T})=\mathbf{j}_{E_2\mathrm{QE}}(\mathbf{R}\hat{T})\,,$$
$$\Lambda_{2ij}(\mathbf{R}\hat{T})=\tfrac{1}{3}\delta_{ij}E_2(\mathbf{R}\hat{T})-\tfrac{3}{4}\tau_{\mathrm{ph}}c^2(\nabla_{\mathbf{R}_i}P_{2j}(\mathbf{R}\hat{T})+\nabla_{\mathbf{R}_j}P_{2i}(\mathbf{R}\hat{T}))+\tfrac{1}{2}\tau_{\mathrm{ph}}c^2\delta_{ij}\nabla_{\mathbf{R}}\delta\mathbf{P}_2(\mathbf{R}\hat{T})\,,\quad(3.26)$$

where

$$\tau_{\mathrm{ph}}=\frac{\beta_0}{\epsilon_{T_0}}\int\frac{d^3p}{(2\pi)^3}\tau(\mathbf{p},\,\omega=cp)c^3p^3\frac{1}{\sinh^2\frac{1}{2}\beta_0cp}\,.\quad(3.27)$$

The function $\tau(\mathbf{p},\omega)$ is the frequency- and momentum-dependent collision time defined by

$$N_1(\mathbf{p}\omega,\mathbf{R}\hat{T})=\tau(\mathbf{p}\omega)N_0(\omega)(1+N_0(\omega))\phi(\mathbf{p}\omega,\mathbf{R}\hat{T})\,,$$

$$\phi(\mathbf{p}\omega,\mathbf{R}\hat{T})=\left(\frac{\beta_0c^2}{4\epsilon_{T_0}}\omega^2\nabla_{\mathbf{R}}\cdot\mathbf{P}_2(\mathbf{R}\hat{T})-\frac{3\beta_0c^2}{4\epsilon_{T_0}}p_ip_j\nabla_{R_i}P_{2j}(\mathbf{R}\hat{T})\right),\quad(3.28)$$

and satisfies as a consequence of the Boltzmann equation

$$\phi(\mathbf{p}\omega,\mathbf{R}\hat{T})=\alpha^2\int\frac{d\omega_1}{2\pi}\int\frac{d^3p_1}{(2\pi)^3}\int\frac{d\omega_2}{2\pi}\int\frac{d^3p_2}{(2\pi)^3}(2\pi)^4\delta(\omega_1-\omega_2-\omega)\delta^3(\mathbf{p}_1-\mathbf{p}_2-\mathbf{p})$$
$$\times[(\omega_1-\omega_2)\mathbf{p}_1\cdot\mathbf{p}_2-\tfrac{1}{2}\omega_1p_2^2+\tfrac{1}{2}\omega_2p_1^2]^2 2\chi_0''(\mathbf{p}_1\omega_1)2\chi_0''(\mathbf{p}_2\omega_2)(N_0(\omega_2)-N_0(\omega_1))$$
$$\times(\tau(\mathbf{p}\omega)\phi(\mathbf{p}\omega,\mathbf{R}\hat{T})+\tau(\mathbf{p}_1\omega_1)\phi(\mathbf{p}_1\omega_1,\mathbf{R}\hat{T})-\tau(\mathbf{p}_1\omega_1)\phi(\mathbf{p}_1\omega_1,\mathbf{R}\hat{T}))\,.\quad(3.29)$$

However, we cannot solve for $\tau(\mathbf{p}\omega)$ and therefore τ_{ph} exactly. We are only able to determine its order of magnitude, for example, by dropping the last two terms in the bracket in (3.29). We find that

$$\tau(\mathbf{p}\omega)\cong(\Sigma'^{>}(\mathbf{p}\omega)-\Sigma'^{<}(\mathbf{p}\omega))^{-1}$$

or

$$\tau(\mathbf{p},\,\omega=cp)\cong(\Sigma'^{>}(\mathbf{p},\,\omega=cp)-\Sigma'^{<}(\mathbf{p},\,\omega=cp))^{-1}\,.\quad(3.30)$$

Since the damping of the phonon is[6]

$$\Gamma(\mathbf{p})=(1/2cp)(\Sigma'^{>}(\mathbf{p},\,\omega=cp)-\Sigma'^{<}(\mathbf{p},\,\omega=cp))\,,\quad(3.31)$$

we have

$$\tau(\mathbf{p},\,\omega=cp)\cong[2cp\Gamma(\mathbf{p})]^{-1}\,.\quad(3.32)$$

The second sound equation can now be obtained by substituting (3.26) into (3.21) and (3.22) and eliminating $\mathbf{P}^{(2)}$. We find

$$\frac{\partial^2}{\partial\hat{T}^2}E_2(\mathbf{R}\hat{T})-\frac{c^2}{3}\nabla_{\mathbf{R}}^2E_2(\mathbf{R}\hat{T})-c^2\tau_{\mathrm{ph}}\nabla_{\mathbf{R}}^2\frac{\partial}{\partial\hat{T}}E_2(\mathbf{R}\hat{T})$$
$$=-\frac{2\alpha}{c^2}\epsilon_{T_0}\left(\frac{\partial^3}{\partial\hat{T}^3}\langle d(\mathbf{R}\hat{T})\rangle-c^2\tau_{\mathrm{ph}}\nabla_{\mathbf{R}}^2\frac{\partial^2}{\partial\hat{T}^2}\langle d(R\hat{T})\rangle\right)$$
$$-\frac{4\alpha}{3c^2}\epsilon_{T_0}c^2\nabla_{\mathbf{R}}^2\frac{\partial}{\partial\hat{T}}\langle d(\mathbf{R}\hat{T})\rangle\quad(3.33)$$

or

$$\frac{\partial^2}{\partial\hat{T}^2}E_2'(\mathbf{R}\hat{T})-\frac{c^2}{3}\nabla_{\mathbf{R}}^2E_2'(\mathbf{R}\hat{T})-c^2\tau_{\mathrm{ph}}\nabla_{\mathbf{R}}^2\frac{\partial}{\partial\hat{T}}E_2'(\mathbf{R}\hat{T})=-\frac{2\alpha}{c^2}\epsilon_{T_0}c^2\nabla_{\mathbf{R}}^2\frac{\partial}{\partial\hat{T}}\langle d(\mathbf{R}\hat{T})\rangle\,,\quad(3.34)$$

where

$$E_2'(\mathbf{R}\hat{T})=E_2(\mathbf{R}\hat{T})+(2\alpha/c^2)\epsilon_{T_0}(\partial/\partial\hat{T})\langle d(\mathbf{R}\hat{T}\rangle\,.\quad(3.35)$$

Equation (3.33) or (3.34) is the same as the phenomenologically derived equation (1.6) for the heat density q when we let $\langle d\rangle=0$ and $\tau_U=\infty$.

IV. LOW-FREQUENCY CORRELATION FUNCTIONS

In this section we will study the two macroscopic equations (2.9) and (3.33) or (3.34) and carry out a hydrodynamic analysis to find the various correlation functions. Our first task is to rewrite (2.9) in a suitable form. Putting $J=0$ and neglecting the $\langle d\rangle^2$ terms, we have

$$(\partial/\partial t_1^2)\langle d(1)\rangle-c^2\nabla_1^2\langle d(1)\rangle+(1/2i)\alpha(123)G(23)=0\,.\quad(4.1)$$

If we use the expression (2.10) for $\alpha(123)$, we find to lowest order in the derivatives $\partial/\partial t_1$ and ∇_1, that

$$\frac{1}{2i}\alpha(123)G(23)=\alpha\left(\frac{1}{2}\frac{\partial}{\partial t_1}\frac{E_1'(\mathbf{r}_1t_1)}{c^2}-\nabla_1\cdot\mathbf{P}_1(\mathbf{r}_1t_1)\right)+\alpha\left(\frac{1}{2}\frac{\partial}{\partial t_1}\frac{E_2(\mathbf{r}_1t_1)}{c^2}-\nabla_1\cdot\mathbf{P}_2(\mathbf{r}_1t_1)\right), \quad (4.2)$$

where $E_1'(\mathbf{r}_1,t_1)$ is a function very similar to $E_1(\mathbf{r}_1,t_1)$:

$$E_1'(\mathbf{r}_1t_1)=\int\frac{d\omega}{2\pi}\int\frac{d^3p}{(2\pi)^3}\times c^2p^2(\chi''(\mathbf{p}\omega,r_1t_1)-\chi_0''(\mathbf{p}\omega))N_0(\omega). \quad (4.3)$$

Now by expressing E_2 and $\mathbf{P}_2$ in terms of E_2', we obtain

$$\frac{1}{2i}\alpha(123)G(23)=\alpha\left(\frac{1}{2}\frac{\partial}{\partial t_1}\frac{E_1'(\mathbf{r}_1t_1)}{c^2}-\nabla_1\cdot\mathbf{P}_1(\mathbf{r}_1t_1)-\frac{\alpha}{c^4}\epsilon_{T_0}\frac{\partial^2}{\partial t_1^2}\langle d(\mathbf{r}_1t_1)\rangle\right)+\frac{3\alpha}{2}\frac{\partial}{\partial t_1}\frac{E_2'(\mathbf{r}_1t_1)}{c^2}. \quad (4.4)$$

The term in the bracket is uninteresting since in our approximation E_1' and P_1 are proportional to $\langle\dot{d}\rangle$ and $\nabla\langle d\rangle$, respectively, and it simply changes the coefficients of $\langle\dot{d}\rangle$ and $c^2\nabla^2\langle d\rangle$ by factors of order α^2. Omitting this term, we have

$$\frac{\partial^2}{\partial t^2}\langle d(\mathbf{r}t)\rangle-c^2\nabla^2\langle d(\mathbf{r}t)\rangle+\frac{3\alpha}{2}\frac{\partial}{\partial t}\frac{E_2'(\mathbf{r}t)}{c^2}=0. \quad (4.5)$$

Before we examine Eqs. (4.5) and (3.34), let us first point out that they can be derived from a much more general basis than our special phonon Hamiltonian with a cubic anharmonic interaction. We begin by noting that $\nabla d(rt)\sim(1/c\sqrt{\rho_0})\mathbf{g}(\mathbf{r}t)$, where ρ_0 is the mean density of the particle system and $\mathbf{g}(\mathbf{r}t)=\rho_0\mathbf{u}(\mathbf{r}t)$ is the particle current density, so that the term $\frac{1}{2}c^2\nabla d\cdot\nabla d$ in the Hamiltonian corresponds to the familiar kinetic term $\frac{1}{2}\rho_0(\mathbf{u})^2$. Then we consider the conservation law for $\mathbf{g}$,

$$\partial g_i/\partial t+\nabla_jT_{ij}=0$$

or

$$(\partial^2/\partial t^2)g_i+\nabla_j(\partial T_{ij}/\partial t)=0, \quad (4.6)$$

where T_{ij} is the stress tensor. Following the procedure described in the Introduction, we expand $\partial T_{ij}/\partial t$ about the local equilibrium value $(\partial T_{ij}/\partial t)_0$

$$\partial T_{ij}/\partial t\cong(\partial T_{ij}/\partial t)_0-\eta\delta_{ij}(\partial/\partial t)\nabla\cdot\mathbf{g}, \quad (4.7)$$

where η is the viscosity. $(\partial T_{ij}/\partial t)_0$ can now be expressed in terms of the independent conserved quantities, which we will choose to be $\mathbf{g}$ and E_2'. We then obtain

$$\left(\frac{\partial T_{ij}}{\partial t}\right)_0=-\delta_{ij}\left(\frac{\partial p}{\partial\rho}\right)_{E_2'}\nabla\cdot\mathbf{g}+\delta_{ij}\left(\frac{\partial p}{\partial E_2'}\right)_{\mathbf{g}}\frac{\partial E_2'}{\partial t}, \quad (4.8)$$

where $p(\mathbf{r}t)$ is the pressure $\frac{1}{3}(T_{ii})_0$. Substituting (4.7) and (4.8) into (4.6) and taking the divergence, we obtain

$$\frac{\partial^2}{\partial t^2}\nabla\cdot\mathbf{g}-\left(\frac{\partial p}{\partial\rho}\right)_{E_2'}\nabla^2(\nabla\cdot\mathbf{g})-\eta\nabla^2\frac{\partial}{\partial t}(\nabla\cdot\mathbf{g})+\left(\frac{\partial p}{\partial E_2'}\right)_{\mathbf{g}}\nabla^2\frac{\partial E_2'}{\partial t}=0. \quad (4.9)$$

This equation is similar to (4.5), except for the term containing η, if we put

$$c^2=(\partial p/\partial\rho)_{E_2'}$$

and

$$\alpha\sim\frac{c}{\sqrt{\rho_0}}\left(\frac{\partial p}{\partial E_2'}\right)_{\mathbf{g}}=\frac{c}{\sqrt{\rho_0}}\left(\frac{\partial p}{\partial T}\right)_{\rho}\frac{1}{c_v}, \quad (4.10)$$

where c_v is the phonon specific heat. The absence of the dissipative term is a result of our not calculating E_1 and $\mathbf{P}_1$ sufficiently accurately. The phenomenological derivation of the equation for E_2' is the same as that for the heat density discussed earlier. The only difference is that there is an extra term that is proportional to $\partial\mathbf{g}/\partial t$ or $(\partial/\partial t)\nabla\langle d\rangle$ in the expansion of $\partial J_{E_2'}/\partial t$, since it is only when the particle current is zero that $\mathbf{j}_{E_2'}$ is equal to $\mathbf{j}_q$. This extra term is just the term $(2\alpha/c^2)\epsilon_{T_0}\times\nabla^2(\partial/\partial t)\langle d\rangle$ in (3.34).

We will now use Eqs. (4.5) and (3.34) to calculate the correlation functions of d and E_2' (denoted by $\langle\mathcal{E}\rangle$ for convenience). The technique of calculation is identical to that described by Kadanoff and Martin.[11] Our notation will also be the same. The underlying idea is to find from these equations the subsequent development of the functions $\langle d\rangle$ and $\langle\mathcal{E}\rangle$ after various adiabatically applied sources are suddenly turned off at $t=0$. We define the one-sided Fourier transforms

$$d(\mathbf{k}z)=\int d^3r\,e^{-i\mathbf{k}\cdot\mathbf{r}}\int_0^\infty dt\,e^{izt}\langle d(\mathbf{r}t)\rangle,$$
$$\mathcal{E}(\mathbf{k}z)=\int d^3r\,e^{-i\mathbf{k}\cdot\mathbf{r}}\int_0^\infty dt\,e^{izt}\langle\mathcal{E}(\mathbf{r}t)\rangle, \quad (4.11)$$

where z is a complex number in the upper half plane. Now taking this transform of Eqs. (4.5) and (3.34), we obtain

$$(-z^2+c^2k^2)d(\mathbf{k}z)-(3\alpha/2c^2)iz\mathcal{E}(\mathbf{k}z)=-izd(\mathbf{k})+(3\alpha/2c^2)\mathcal{E}(\mathbf{k}), \quad (4.12)$$

$$(-z^2+\tfrac{1}{3}c^2k^2-iz\tau_{\rm ph}c^2k^2)\mathcal{E}(\mathbf{k}z)-(2\alpha/c^2)\epsilon_{T_0}izc^2k^2d(\mathbf{k}z)=(-iz+\tau_{\rm ph}c^2k^2)\mathcal{E}(\mathbf{k})+(2\alpha/c^2)\epsilon_{T_0}c^2k^2d(\mathbf{k}). \quad (4.13)$$

The functions $d(\mathbf{k})$ and $\mathcal{E}(\mathbf{k})$ are the spatial transforms

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[11] L. Kadanoff and P. C. Martin, Ann. Phys. (N. Y.) **24**, 419 (1963).

of $\langle d\rangle$ and $\langle\mathcal{E}\rangle$ at $t=0$:

$$d(\mathbf{k})=\int d^3r\, e^{-i\mathbf{k}\cdot\mathbf{r}}\langle d(\mathbf{r},0)\rangle\,,$$
$$\mathcal{E}(\mathbf{k})=\int d^3r\, e^{-i\mathbf{k}\cdot\mathbf{r}}\langle \mathcal{E}(\mathbf{r},0)\rangle\,. \tag{4.14}$$

We have also used the initial condition that $(\partial/\partial t)\langle d(\mathbf{r}t)\rangle$ and $(\partial/\partial t)\langle\mathcal{E}(\mathbf{r}t)\rangle$ at $t=0$ are zero. We may determine by perturbation theory the values of $d(\mathbf{k})$ and $\mathcal{E}(\mathbf{k})$ induced by sources externally applied for $t<0$ according to the Hamiltonian

$$\mathcal{H}(\mathbf{r})=\mathcal{H}_0(\mathbf{r})-(d(\mathbf{r})J(\mathbf{r})+[\delta T(\mathbf{r})/T_0]\mathcal{H}_0(\mathbf{r}))\times\eta(-t)e^{\epsilon t}\,, \tag{4.15}$$

$[\eta(x)=1$ for $x>0$ and $\eta(x)=0$ for $x<0$, and $\mathcal{H}_0(\mathbf{r})$ is the unperturbed Hamiltonian density$]$:

$$\langle d(\mathbf{r},0)\rangle=\frac{\mathrm{Tr}[\exp(-\beta_0\int d^3r\,\mathcal{H}(\mathbf{r}))d(\mathbf{r})]}{\mathrm{Tr}[\exp(-\beta_0\int d^3r\,\mathcal{H}(\mathbf{r}))]}\,,$$
$$\langle \mathcal{E}(\mathbf{r},0)\rangle=\frac{\mathrm{Tr}[\exp(-\beta_0\int d^3r\,\mathcal{H}(\mathbf{r}))\mathcal{H}_0(\mathbf{r})]}{\mathrm{Tr}[\exp(-\beta_0\int d^3r\,\mathcal{H}(\mathbf{r}))]}-\epsilon_0\,. \tag{4.16}$$

In obtaining the latter expression, we have recalled the fact that, when $\langle\dot{d}\rangle=0$, $\langle\mathcal{E}\rangle$ is simply E_2, the thermal energy of the phonons, so that the zero-point energy is subtracted out. If we express $d(\mathbf{k})$ and $\mathcal{E}(\mathbf{k})$ in terms of $J(\mathbf{k})$ and $\delta T(\mathbf{k})$ and note that

$$\frac{\delta d(\mathbf{k})}{\delta J(\mathbf{k})}\to\frac{1}{c^2k^2};\quad \frac{\delta\mathcal{E}(\mathbf{k})}{\delta T(\mathbf{k})}\to c_v\,,$$
$$\frac{\delta d(\mathbf{k})}{\delta T(\mathbf{k})}\to 0;\quad \frac{\delta\mathcal{E}(\mathbf{k})}{\delta J(\mathbf{k})}\to 0\,, \tag{4.17}$$

we find

$$(-z^2+c^2k^2)d(\mathbf{k}z)-(3\alpha/2c^2)iz\mathcal{E}(\mathbf{k}z)=-(iz/c^2k^2)J(\mathbf{k})+(3\alpha/2c^2)c_v\delta T(\mathbf{k})\,, \tag{4.12$'$}$$

$$(-z^2+\tfrac{1}{3}c^2k^2-iz\tau_{\mathrm{ph}}c^2k^2)\mathcal{E}(\mathbf{k}z)+(2\alpha/c^2)\epsilon_{T_0}izc^2k^2d(\mathbf{k}z)=(-iz+\tau_{\mathrm{ph}}c^2k^2)c_v\delta T(\mathbf{k})-(2\alpha/c^2)\epsilon_{T_0}J(\mathbf{k})\,. \tag{4.13$'$}$$

These equations allow us to calculate the Fourier transforms of the response functions, or retarded commutators,[11] of the operators $d(\mathbf{r}t)$ and $\mathcal{E}(\mathbf{r}t)$:

$$\chi_{AB}(\mathbf{k}z)=\int d^3r\, e^{-i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}\int_{-\infty}^{\infty}dt\, e^{iz(t-t')}\times i\eta(t-t')\langle[A(\mathbf{r}t),B(\mathbf{r}'t')]\rangle\,. \tag{4.18}$$

They reduce to

$$\chi_{dd}(\mathbf{k}z)=\frac{-z^2+\frac{1}{3}c^2k^2}{\{(z^2-c^2k^2)[z^2-\frac{1}{3}c^2k^2]-(3\alpha^2/c^4)\epsilon_{T_0}z^2c^2k^2\}}\,,$$
$$\chi_{d\mathcal{E}}(\mathbf{k}z)=\frac{(\alpha/2c^2)T_0c_vizc^2k^2}{\{(z^2-c^2k^2)[z^2-\frac{1}{3}c^2k^2]-(3\alpha^2/c^4)\epsilon_{T_0}z^2c^2k^2\}}\,,$$
$$\chi_{\mathcal{E}\mathcal{E}}(\mathbf{k}z)=\frac{\frac{1}{3}T_0c_vc^2k^2(-z^2+c^2k^2)}{\{(z^2-c^2k^2)[z^2-\frac{1}{3}c^2k^2]-(3\alpha^2/c^4)\epsilon_{T_0}z^2c^2k^2\}}\,,$$
$$\chi_{\mathcal{E}d}(\mathbf{k}z)=\frac{-(2\alpha/c^2)\epsilon_{T_0}izc^2k^2}{\{(z^2-c^2k^2)[z^2-\frac{1}{3}c^2k^2]-(3\alpha^2/c^4)\epsilon_{T_0}z^2c^2k^2\}}\,, \tag{4.19}$$

when we neglect dissipative terms containing τ_{ph}, which are smaller by a factor of $z\tau_{\mathrm{ph}}$. We will come back to these terms in the next section where χ_{dd} is rederived in a slightly different way. We note that $\chi_{\mathcal{E}d}=-\chi_{d\mathcal{E}}$, as it must, since for harmonic phonons, $\frac{1}{2}T_0c_v=\frac{1}{2}T_0(\partial\epsilon_{T_0}/\partial T_0)_d$ is equal to $2\epsilon_{T_0}$.

These response functions coincide with those obtained from the two-fluid hydrodynamic analysis carried out by Hohenberg and Martin,[4] if we identify

$$(3\alpha^2/c^4)\epsilon_{T_0}=((c_p/c_v)-1)\,, \tag{4.20}$$

which agrees with (4.11) upon using

$$\left(\frac{c_p}{c_v}-1\right)=-\frac{T}{c_v\rho_0}\left(\frac{\partial p}{\partial T}\right)_\rho^2\bigg/\left(\frac{\partial p}{\partial\rho}\right)_T\,.$$

We may rewrite these functions to exhibit the two renormalized sound poles with velocities u_1 and u_2 defined by

$$(z^2-u_1{}^2k^2)(z^2-u_2{}^2k^2)=(z^2-c^2k^2)(z^2-\tfrac{1}{3}c^2k^2)-(c_p/c_v-1)z^2c^2k^2$$

or

$$u_1{}^2+u_2{}^2=[\tfrac{4}{3}+((c_p/c_v)-1)]c^2;\quad u_1{}^2u_2{}^2=\tfrac{1}{3}c^4\,. \tag{4.21}$$

In particular, when $[(c_p/c_v)-1]$ is much smaller than one, a condition that is certainly satisfied at sufficiently low temperature, we have

$$\chi_{dd}(\mathbf{k}z)=\frac{1-\frac{3}{4}((c_p/c_v)-1)}{-z^2+u_1{}^2k^2}+\frac{\frac{3}{4}((c_p/c_v)-1)}{-z^2+u_2{}^2k^2}\,,$$
$$\chi_{d\mathcal{E}}(\mathbf{k}z)=-\frac{3\alpha}{4c^2}T_0c_viz\left(\frac{1}{-z^2+u_1{}^2k^2}-\frac{1}{-z^2+u_2{}^2k^2}\right),$$
$$\chi_{\mathcal{E}\mathcal{E}}(\mathbf{k}z)=\tfrac{1}{3}T_0c_vc^2k^2\left(\frac{9/4((c_p/c_v)-1)}{-z^2+u_1{}^2k^2}+\frac{1-9/4((c_p/c_v)-1)}{-z^2+u_2{}^2k^2}\right), \tag{4.22}$$

where[12]

$$u_1^2 \cong c^2 + \tfrac{3}{2}((c_p/c_v)-1)c^2\,,$$
$$u_2^2 \cong \tfrac{1}{3}c^2 - \tfrac{1}{2}((c_p/c_v)-1)c^2\,. \tag{4.23}$$

V. ALTERNATIVE DERIVATION OF THE DISPLACEMENT CORRELATION FUNCTION

In this section, we will rederive $\chi_{dd}(\mathbf{k}z)$ using another approach. First of all, it can be easily shown that $\chi_{dd}(\mathbf{k}z)$ is simply the analytic continuation of the function[8]

$$G(\mathbf{k}\omega_\nu) = \int d^3r\, e^{-i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} \int_0^{-i\beta} dt\, e^{+i\omega_\nu(t-t')} G(\mathbf{r}t,\mathbf{r}'t'),$$
$$\omega_\nu = 2\pi\nu/-i\beta;\quad \nu = \text{integer}\,, \tag{5.1}$$

which is the Fourier transfer of the imaginary-time Green's function, from ω_ν to z in the upper half-plane. The expression for χ_{dd} in the hydrodynamic limit shown in (4.22) can therefore be obtained alternatively by calculating $G(\mathbf{k}\omega_\nu)$ or $G(\mathbf{k},z)$ at small momentum $\mathbf{k}$ and frequency ω_ν.

From (2.11) and (2.12), we find that

$$G(\mathbf{k}\omega_\nu) = [-\omega_\nu^2 + c^2k^2 - \Sigma(\mathbf{k}\omega_\nu)]^{-1}\,, \tag{5.2}$$

where

$$\Sigma(\mathbf{k}\omega_\nu) = \int d^3r\, e^{-i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} \int_0^{-i\beta} dt\, e^{+i\omega_\nu(t-t')} \times\left[-\frac{1}{2i}\alpha(123)\left(\frac{\delta G(23)}{\delta\langle d(1')\rangle}\right)_{\langle d\rangle\to 0}\right]. \tag{5.3}$$

Since we only need $\Sigma(\mathbf{k}\omega_\nu)$ at low $\mathbf{k}$ and ω_ν, we can make use of the expansion (4.2) or (4.4) for $(1/2i)\alpha(123)G(23)$, getting

$$\Sigma(\mathbf{k}\omega_\nu) = \int d^3r\, e^{-i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} \int_0^{-i\beta} dt\, e^{+i\omega_\nu(t-t')} \times\left(-\frac{3\alpha}{2c^2}\frac{\delta}{\delta\langle d(1')\rangle}\frac{\partial}{\partial t}E_2'(\mathbf{r}t)\right), \tag{5.4}$$

where $E_2'(\mathbf{r}t)$ is the previously defined function except that t is in the imaginary time interval $\{0, -i\beta\}$. We have neglected the terms depending on $\langle d\rangle$ explicitly. $\Sigma(\mathbf{k}\omega)$ can now be evaluated by taking the functional derivative on the equation for $E_2'(\mathbf{r}t)$ (3.34), which is also valid for imaginary times. The result for the continuation is

$$\Sigma(\mathbf{k}\omega) \cong \frac{(3\alpha^2/c^4)\epsilon_{T_0}\omega^2c^2k^2}{-\omega^2+\frac{1}{3}c^2k^2 - i\omega\tau_{\text{ph}}c^2k^2}$$

or

$$= \frac{((c_p/c_v)-1)\omega^2c^2k^2}{-\omega^2+\frac{1}{3}c^2k^2 - i\omega\tau_{\text{ph}}c^2k^2}\,, \tag{5.5}$$

using (4.20). Thus

$$G(\mathbf{k}\omega) = [-\omega^2 + c^2k^2 - ((c_p/c_v)-1)\omega^2c^2k^2/(-\omega^2+\tfrac{1}{3}c^2k^2 - i\omega\tau_{\text{ph}}c^2k^2)]^{-1}, \tag{5.6}$$

which is identical to our previous result for χ_{dd}, if we put $\omega\tau_{\text{ph}}=0$.

Let us now discuss the implication of this formula. As is well known, the complex poles of the retarded Green's function represent the frequency and damping of the normal modes or elementary excitations of the system, which are, in our case, the excitations of the atomic vibrations or density fluctuations. From (5.6), we find that $G(\mathbf{k}\omega)$ can be expressed as a sum of two poles. In particular, for small $[(c_p/c_v)-1]$ and $\omega\tau_{\text{ph}}$, we find

$$G(\mathbf{k}\omega) = \frac{-[1-\frac{3}{4}((c_p/c_v)-1)]}{-\omega^2 - i\omega\tau_1c^2k^2 - u_1^2k^2} + \frac{-\frac{3}{4}((c_p/c_v)-1)}{-\omega^2 - i\omega\tau_2c^2k^2 - u_2^2k^2}\,, \tag{5.7}$$

where

$$\tau_2 = \frac{9}{4}\left(\frac{c_p}{c_v}-1\right)\tau_{\text{ph}}\,,$$
$$\tau_2 = \left[1-\frac{9}{4}\left(\frac{c_p}{c_v}-1\right)\right]\tau_{\text{ph}}\,, \tag{5.8}$$

and u_1^2 and u_2^2 are given by (4.21) or (4.23). The first term describes the first sound with velocity u_1 and damping proportional to $\omega^2\tau_1$. The other term corresponds to the second sound with velocity u_2 and damping $\omega^2\tau_2$. Since $[(c_p/c_v)-1]$ is very small, the damping of the first sound is much smaller than that of the second sound, even though they are both described by the relaxation time τ_{ph}. It is obvious from (5.7) that the second sound mode must be treated like the first sound mode, since they appear on the same footing as elementary excitations of the system. In other words, both of them are phonons. The only difference is that the second sound appears with a weight smaller by $[(c_p/c_v)-1]$.

[12] We note that they are the two fluid analog of the equations $u_1^2 \cong c^2 + (c_p/c_v - 1)c^2$; $u_2^2 = 0$, which give the adiabatic compressibility for the sound velocity when there is no second sound.