The role of long-wave longitudinal phonons in nonlocal heat conduction

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Heat transport in “dirty” dielectrics, in which thermal phonons with \( \omega \ll T \) are more strongly scattered by static defects than by one another, has been studied. Under these conditions, as has already been shown [R. P. B. A. Landau, Zh. Eksp. Teor. Fiz. 82, 1936–1944 (June 1982)], it was considered that the sole dissipative mechanism for the momentum transfer of energy through the phonon spectrum. Since the properties of longitudinal and transverse subthermal phonons are very different, the existence of various branches of the spectrum changes the phonon kinetics and has a strong influence on the nature of the heat conduction.

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It was shown previously⁵ that thermal conductivity resulting from Rayleigh scattering of phonons by defects is highly specific; it is nonlocal and is connected with the transfer of energy through the spectrum (from the region of thermal frequencies \( \omega \ll T \), where the energy is stored, to regions of low subthermal frequencies \( \omega = T \) where it is transported spatially). Two assumptions were made there: it was considered that the kinetic equation for \( \omega \) phonons could be replaced by a diffusion equation and that all phonons belong to a single isotropic branch which was such that \( \omega \) phonons can be scattered by thermal phonons. Both these assumptions are only valid for sufficiently “dirty” crystals, when scattering by defects predominates over phonon-phonon scattering. Besides, with a different form of averaging, for example substituting \((\omega - \omega_0) \sim 1\) for an expression in terms of \( \omega_0 \), the divergence disappears. This shows that it is preferable to evaluate \( \omega \) not by resorting to an a priori approximation scheme, but by starting from the kinetic equation. It should also be pointed out that scattering by defects with mode conversion was not taken into account in the works quoted above. Yet such processes, which transform into one another subthermal phonons having different behaviors with respect to phonon-phonon interactions, are extremely important.

1. STATEMENT OF THE PROBLEM

We consider an unbounded dielectric at low temperatures, when umklapp processes can be neglected, so that the sole dissipative mechanism for the momentum of the phonon system is Rayleigh scattering by defects. We assume that the departure of the crystal from thermodynamic equilibrium (with temperature \( T_0 \)) is small and can be described by the local temperature \( T = T_0 + \delta T(r, t) \). The small departure from equilibrium implies that the phonon distribution is of the form

\[
\rho_n(q, \omega, t) = n_0(\omega) + \delta n_0(q, \omega, t),
\]

where \( \delta n_0 \) is small. Here \( q \) is the phonon momentum, \( \omega \) its polarization, and \( n_0 \) the Planck distribution at temperature \( T_0 \).

On the assumption that the energy flow \( w \) is borne by subthermal phonons with \( \omega \ll T_0 \), the anisotropic part \( \delta m^a \) of the correction \( \delta m \) can be neglected; on the other hand, the assumption of the existence of a local temperature implies that the isotropic part of this correction, \( \delta m^a \) has at \( \omega = T_0 \) the form

\[
\delta m^a = \text{constant.}
\]
where the order of unity, \( a \), and respectively, linearized in collision terms for defects and three-phonon processes for subthermal longitudinal phonons take place appreciably more slowly than for transverse phonons. In addition, \( S_j \) unlike \( S_0 \) does not contain \( \delta T \) [the collision integral Eq. (9) does not lead to the establishment of equilibrium].

If we go over to an isotropic model of the spectrum, then the form of \( S_j \) does not change considerably; the difference is just that \( b \) depends on \( e \). However, \( S_j \) can change appreciably on going over to an anisotropic model. In particular, thanks to the anisotropy, the prohibition on the interaction between longitudinal and thermal phonons can be lifted. \( S_j \) will then have a form analogous to Eq. (7), namely

\[
S_j(\omega, e) = -\frac{1}{v_0(e, \omega)} \left[ \delta T(\omega, e) - \delta T \right],
\]

where the exponent \( m \) depends on the type of crystal.\(^1\) It is important, however, that \( m > 2 \); in the anisotropic, as in the isotropic model. Therefore, phonon-phonon processes for longitudinal phonons go more slowly than for transverse phonons (although the difference may not be so great).

Substituting the expressions for \( S_j \) and \( S_0 \) into Eq. (4) we obtain a system in which the terms in \( \delta T \) must be regarded as inhomogeneities and a solution of proportional to \( \delta T \) must be found. Having found \( \delta n_\omega \), the current can be calculated

\[
\frac{1}{v_0(e, \omega)} - \frac{1}{v_0(e, \omega)} \frac{\partial n_\omega}{\partial T}.
\]

The phonon-phonon interaction terms are very different for transverse and longitudinal phonons. We shall first examine the isotropic model. Transverse subthermal phonons interact mainly with thermal phonons; using Eq. (2) in the linearization, we can obtain for \( \omega \ll T \),

\[
S_j(\omega, e) = -\frac{1}{v_0(e, \omega)} \left[ \delta T(\omega, e) - \delta T \right].
\]

The time \( \tau_j(\omega) \) here is the time for attestation of transversal sound; it is simultaneously the time for establishing the Planck equilibrium in the low-frequency region. It can be written in the following form:

\[
\frac{1}{v_0(e, \omega)} - \frac{1}{v_0(e, \omega)} \frac{\partial n_\omega}{\partial T}, \quad b=1.
\]

Interaction between subthermal longitudinal phonons and thermal phonons is forbidden by the energy and momentum conservation laws, so that \( S_j \) has a different form:

\[
S_j(\omega, e) = -\frac{1}{v_0(e, \omega)} \left[ \delta T(\omega, e) - \delta T \right]
\]

The main frequency dependence enters in the longitudinal-sound attenuation time, which can conveniently be written in the form:

\[
\frac{1}{v_0(e, \omega)} - \frac{1}{v_0(e, \omega)} \frac{\partial n_\omega}{\partial T}, \quad b=1
\]

while the functions \( S_j \) of the order of unity) are so constructed that in integrating over frequency we have \( \omega \)
$D_0 = 1/3 \nu_0^2 T$ is the diffusion coefficient for thermal phonons. It is not necessary to distinguish between longitudinal and transverse phonons in the parameters $\tau$, and $I$, since for thermal phonons all differences reduce to factors of the order of unity. By evaluating the current $\bar{w}$, it can be seen which phonons $\omega$ are important in energy transport. The assumptions made above are valid if

$$a \approx xT.$$  \hspace{1cm} (16)

When solving the system of Eq. (4) it can be considered, as before,1 that energy transport takes place quasistationarily, i.e., that the distribution function $f_0$ in the low-frequency region follows a slowly varying temperature $\delta T$. The justification for this assumption is the inequality

$$\tau \approx \tau_\text{SF} \ll 1.$$  \hspace{1cm} (17)

It is convenient to solve Eq. (4) by making the Fourier transformation $\tau \rightarrow \omega$, after which it takes the form

$$\delta \tau_\omega = -i \omega \delta T.$$  \hspace{1cm} (18)

We write the Fourier component of the energy flux in the form

$$\bar{w} = -i \omega \bar{\tau}\delta T.$$  \hspace{1cm} (19)

If the heat conduction is nonlocal, $\bar{\tau}$ depends on $\omega$.

2. SEMIQUANTITATIVE ANALYSIS OF THE KINETIC EQUATION

We use the following means to assess range of the parameters in which various collision and drift terms are important. We choose Eq. (11) for $S_1$ and neglect all angular dependences of the coefficients $a$ and $b$ in Eqs. (5) and (11). The $\delta \tau_0$ from Eq. (4) can be expressed linearly in terms of $\delta T$ and of the isotropic parts of the corrections $\delta \omega_i$. By integrating these relations over the angles, we obtain a system of linear equations for $\delta \omega_i$ and an expression for $\delta \omega_i$ in terms of $\delta T$. Substituting them into the above expression for $\delta \tau_0$, in terms of $\delta \omega_i$ and $\delta T$, we finally obtain $\delta \tau_0$ proportional to $\delta T$. Knowing $\delta \tau_0$, we can evaluate the fluxes $\bar{w}$, and find which frequencies and polarizations transport energy, thereby verifying the inequality Eq. (16). Knowing the fluxes, the characteristic time $\tau$ for processes can be found and inequalities (15) and (17) verified. As such an analysis shows, the simultaneous satisfaction of inequalities (15) and (16) imposes a definite limit on $L \ll \nu^*$. It is convenient to represent it in the plane of the parameters $L/\nu_0$ and $\delta \omega_0/\nu_0^2$ (see Fig. 2 in Levinson1). The form of the region in which inequalities (15) and (16) are satisfied depends on how effective phonon-phonon processes are for longitudinal phonons. In the isotropic model, which corresponds to $m=4$, the region sought is bounded by the inequalities

$$L/\nu_0 > \frac{1}{4} \delta \omega_0,$$

$$L/\nu_0 > \frac{1}{8} \delta \omega_0.$$  \hspace{1cm} (20)

It is situated to the right of the thick lines in Fig. 1(a). In cubic crystals, which correspond to $m=3$, and where phonon-phonon processes for longitudinal phonons are stronger, the region where subthermal phonons are important is smaller [the region to the right of the thick lines in Fig. 1(16)]. It is bounded by the inequalities

$$L/\nu_0 > \frac{1}{3} \delta \omega_0.$$  \hspace{1cm} (21)

The thin lines in the figure show the subdivisions of the regions in which different terms of the kinetic equation are important and in which the thermal conductivity has different dependences on $k$.

For both the isotropic model and for a cubic crystal there are three regions, of which region I coincides with the region studied before.1 In region I longitudinal and transverse phonons give identical contributions to the current, and the frequencies of interest $\omega_i = \omega_{i0} = \omega$ are such that for them

$$D(\omega_i) = \omega^4,$$  \hspace{1cm} (22)

i.e., the diffusion length during the time for attenuation by the "thermostat" of thermal phonons is of the order of the dimensions of the disturbed region. The times for the frequencies of interest are in the order $\tau(\omega_i) = \tau(\omega_{i0}) + \tau(\omega)$. The coefficient of thermal conductivity is

$$\kappa(\omega_i) = \kappa(\omega_{i0}) + \kappa(\omega),$$  \hspace{1cm} (23)

where $\kappa_i$ is a numerical coefficient of order unity, $\kappa_{00} = \kappa_0 \tau_0^4 = \kappa_0 \tau_0^4$ is the "nominal" thermal conductivity, and $\tau = \nu T^3$. The result in Eq. (24) is the same as that obtained earlier1 where one phonon branch with the properties of transverse phonons was studied. This is not surprising since the fastest processes in region I are scattering by defects, and mix not only the direction of propagation but also the polarization. Longitudinal phonons in region I relax to equilibrium by conversion into transverse through scattering by defects. The term $S_2$ can therefore be discarded and its choice in the form of Eq. (11) rather than Eq. (9) has no effect. As we will show below (taking other regions as examples), the angular dependence of the coefficients $a$ and $b$ only affects the magnitude of the coefficient $\kappa_i$.

In regions II and III, i.e., in purer crystals, the current is determined only by longitudinal phonons, for which it is easier to destroy the quasi-equilibrium distribution, since anharmonic processes are weaker for them. Transverse phonons in regions II and III can be

FIG. 1. Regions for values of the parameters where the energy is transported by subthermal phonons: (a) isotropic medium; (b) a cubic $\bar{f}$-class crystal. The shaded area shows where heat conduction is nonlocal.
regarded as being in quasi-equilibrium, i.e.,
\[ \bar{v}_n = \bar{v}_0 T, \quad \bar{v}_0 = \text{const}. \]  
(25)
Transverse phonons form a thermostat for longitudinal phonons; there are two interaction mechanisms between the longitudinal-phonon system and the transverse-phonon system: scattering by defects with mode conversion and phonon-phonon interactions. In region II the energy is transported by transverse phonons with a mean free path for impurities of the order of the macroscopic dimensions:
\[ \bar{v}_x = \bar{v}_0 T \]  
(26)
Here
\[ \bar{v}_x = \bar{v}_x (\bar{v}_0 T), \]  
(27)
so that the interaction of longitudinal phonons with the transverse-phonon thermostat takes place by mode conversion on scattering by defects. The term \( \bar{v}_x \) must be accounted for in region I, and the difference between an isotropic medium and a cubic crystal reduces to negligible angular dependences of the coefficients \( \bar{v}_x \) and \( \bar{v}_0 \). The thermal conductivity is determined by the expression
\[ \bar{\kappa} = \bar{v}_x / \bar{v}_0 T \]  
(28)
where \( N \) is the defect concentration; the thermal conductivity [Eq. (28)] does not depend on \( T \).

The collision term \( \bar{v}_c \) must be taken into account in region III so that the properties of an isotropic medium and of a cubic crystal are different. In the first case longitudinal phonons carry the current and their mean free path for phonon-phonon scattering is of the order of \( L \): 
\[ \bar{v}_c = \bar{v}_c (\bar{v}_0 T) \]  
(29)
The inequality (27) is then reversed:
\[ \bar{v}_c = \bar{v}_c (\bar{v}_0 T) \]  
(30)
and the thermal conductivity
\[ \bar{\kappa} = \bar{\kappa} - \bar{\kappa} \bar{v}_c / \bar{v}_0 T \]  
(31)
does not depend on the defect concentration since the interaction between longitudinal phonons and the thermostat of the transverse phonons is entirely determined by phonon-phonon scattering. This agrees with the fact that for an isotropic medium region III corresponds to pure crystals with \( b \ll 1 \). The nonlocal conductivity (31) becomes equal on the left-hand boundary of region III to the usual local thermal conductivity
\[ \bar{\kappa} = \bar{\kappa} - \bar{\kappa} \bar{v}_c / \bar{v}_0 T \]  
(32)
which obtains in pure specimens where a displaced Planck distribution is established and the thermal conductivity is determined by the mean (1/\( T \)) and not by the mean (\( \bar{v}_c \)).

In a cubic crystal the energy is transported by longitudinal phonons in region III for which
\[ \bar{v}_x = \bar{v}_x (\bar{v}_0 T) \]  
(33)
I.e., they have a noticeable probability of being scattered by transverse phonons. Then

This thermal conductivity is local but it depends not only on \( T \) but also on \( \bar{v}_x \). This means that spectral transport takes place in the process of heat transport, which is unavoidable if subthermal phonons transport the energy. At the left-hand boundary of region III the thermal conductivity of Eq. (34) joins to Eq. (32).

Equation (28) for the thermal conductivity in region III for an isotropic medium agrees with Pomeranchuk's results. In this connection we note that on the low temperature side or for "cleaner" crystals the Casimir region of thermal conductivity, \( \bar{\kappa} - \bar{\kappa} \bar{v}_c / \bar{v}_0 T \) [Eq. (32)] borders on region II in Pomeranchuk's work (where the mean free path of longitudinal phonons is limited by the specimen dimensions) and not region I with thermal conductivity (28). This comes about by Pomeranchuk's neglect of scattering by defects with conversion of longitudinal into transverse phonons.

3. ALLOWANCE FOR THE EXACT FORM OF THE COLLISION TERMS

We now show that taking account of the angular dependences of the coefficients \( \bar{v}_x \) and \( \bar{v}_0 \) and also the choice of the correct form of the collision term \( \bar{v}_c \), do not influence the values and power dependences given in the previous section. We first carry this out taking as an example region III for an isotropic medium. We suppose, as in the semiquantitative analysis, that impurities are unimportant and that transverse phonons are in quasi-equilibrium. Then, substituting Eq. (23) into Eq. (9), we obtain the following equation:
\[ \frac{\partial T}{\partial \bar{v}_x} \frac{\partial \bar{v}_c}{\partial \bar{v}_0 T} + \frac{\partial \bar{v}_c}{\partial \bar{v}_0 T} \frac{\partial T}{\partial \bar{v}_x} = \int \frac{\partial \bar{v}_c}{\partial \bar{v}_x} \frac{\partial \bar{v}_c}{\partial \bar{v}_0 T} \frac{\partial T}{\partial \bar{v}_x} \]  
(35)
where \( \theta \) is the angle between \( \bar{v}_x \) and \( \bar{v}_0 \), and
\[ \bar{v}_0 = \int \frac{\partial \bar{v}_c}{\partial \bar{v}_x} \frac{\partial \bar{v}_c}{\partial \bar{v}_0 T} \frac{\partial T}{\partial \bar{v}_x} \]  
(36)
All the important parameters can be eliminated from this equation; for this purpose Eq. (10) must be substituted into it and
\[ \bar{v}_c (\bar{v}_0 T) = \bar{v}_0 \bar{v}_0 T \]  
(37)
We then obtain for \( \bar{v}_x \) the equation
\[ \int \frac{\partial \bar{v}_c}{\partial \bar{v}_x} \frac{\partial \bar{v}_c}{\partial \bar{v}_0 T} \frac{\partial T}{\partial \bar{v}_x} = - \int \frac{\partial \bar{v}_c}{\partial \bar{v}_x} \frac{\partial \bar{v}_c}{\partial \bar{v}_0 T} \frac{\partial T}{\partial \bar{v}_x} = 0. \]  
(38)
All the parameters in it are of the order of unity; so it is the function \( F \). Calculating the flux \( \bar{v}_0 \) with the help of the distribution (37), we come to Eq. (31), where \( \bar{v}_c \) is some integral of \( F \). The use of the correct collision term \( \bar{v}_c \) in the form (9) and not in the form of (11), as in the semiquantitative analysis of the kinetic equation, had thus no important influence on the results.

We now consider a cubic crystal. In agreement with the results of the semiquantitative analysis, we can take

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\[ \delta n(v) = \delta T/\omega L \delta n'(v) \delta n'(0), \]  
(39)

where the anisotropic addition is small. If such a distribution and Eq. (25) are substituted into \( \delta n \), all terms in \( \delta T \) cancel. The same takes place on substitution into \( \delta T \), which must now be chosen in the form of Eq. (11).

As a result we obtain the equation

\[ \delta n'(v) = -\frac{\delta T}{\omega} \delta n'(0), \]  
(40)

It can already be seen from this that the current is local, since \( \delta n' \) is proportional to \( b \). To separate the dependence of current on \( \delta n \), we make the substitution

\[ \delta n'(v) = -\frac{\delta T}{\omega} G(v), \]  
(41)

and substitute Eqs. (12) with \( m = 2 \) and Eq. (6) in Eq. (40). It is easy to confirm that the equation obtained for \( F \) contains only parameters of order unity and that the flux (34) is obtained from the distribution (41).

4. THE ROLE OF HIGHER-ORDER ANHARMONIC PROCESSES

The prohibition of interaction between subthermal longitudinal phonons with thermal phonons can even be lifted within the framework of the isotropic model. For this, either four-phonon processes\(^1\) must be taken into account, or else it must be assumed that in a three-phonon almost collinear process in which three longitudinal phonons take part the energy conservation law is relaxed because of the finite lifetime of a thermal phonon (the Simons mechanism\(^2\)). Such mechanisms naturally lead to a local thermal conductivity and this was calculated by Gurzhi and Maksimov\(^3\) for the Simons mechanism. They are important for large dimensions \( L \) and relatively high temperatures \( T \). Appropriate criteria are not difficult to write down for all the regions considered, if it is taken into account that the time for four-phonon processes for subthermal phonons is calculated in the following way:\(^4\)

\[ \tau = \gamma \omega_0 / \omega_0^2 \tau_0^2 \]  
(42)

where \( \omega_0 \) is the Debye frequency and \( \gamma = \omega_0 / M^2 \) is the square of the adiabatic parameter \( (M \) is the mass of a unit cell), while the prohibition on three-phonon processes is completely lifted for\(^5\)

\[ \omega_0^2 < \omega_0^2 \tau_0 / \gamma \]  
(43)