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LETTER TO THE EDITOR

ON THE LATTICE THERMOCONDUCTIVITY OF AN IDEAL CRYSTAL

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The lattice thermoconductivity of an ideal crystal is obtained as a function of temperature by using a radiation-like description of the heat carriers (Debye phonons).

It is well-known from classical arguments[1)] that the lattice thermoconductivity of an ideal crystal as a function of the temperature T behaves like T^3 at low temperatures (boundary scattering limit)[2)] and obeys a $1/T$-law in the high-temperature limit[3)] with an in-between peak which is usually referred to as the umklapp-scattering contribution[4)]. This behaviour is obtained in the present paper by computing the lattice thermoconductivity of an ideal crystal in the framework of the radiation-like description of the Debye phonons. In addition, the thermoconductivity corresponding to the non-equilibrium transport of energy is obtained, which behaves like $1/T^2$ in the high-temperature limit. The radiation like description of the phonons is based on the natural assumption that the carriers are absorbed, during the energy transfer process, at a rate which is proportional to their thermal occupation number. The standpoint expressed by Peierls[4)] is taken as regards the ideal crystal, in that the phonons are viewed as well-defined elementary excitations. The effect of various collision processes is shown how to be included in the present approach.

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As in the usual experiments of measuring the thermoconductivity, consider a rod of an ideal crystal, oriented along the z-axis and having a length much greater than its transverse size d. Under these circumstances one assumes that the thermal equilibrium is attained. A slight temperature difference ΔT is overimposed along the rod, such that the heat flows only along the z-axis. The $\vec{q}$-th mode in a phonon branch with the frequency $\omega(\vec{q})$ contributes $(\partial\omega(\vec{q})n(\vec{q}))/\partial T)\Delta T$ to the density of the flowing heat, where $n(\vec{q})$ is the Planck–Einstein distribution. This energy is carried along the z-axis with the group velocity

$$v_z(\vec{q}) = \partial\omega(\vec{q})/\partial q_z; \tag{1}$$

therefore, the average heat flux along this axis is

$$Q(\vec{q}) = \frac{\partial}{\partial T}\big[\omega(\vec{q})n(\vec{q})\big] \cdot v_z(\vec{q})\tau_z(\vec{q}) \cdot \Delta T\,, \tag{2}$$

where $\tau_z(\vec{q})$ is the corresponding relaxation time. The total flow of heat (heat current) is obtained from the continuity equation and by summing over all the phonon modes,

$$\frac{\partial Q}{\partial t} = \frac{1}{(2\pi)^3}\frac{\partial}{\partial z}\left\{\int \mathrm{d}\vec{q}\frac{\partial}{\partial T}\big[\omega(\vec{q})n(\vec{q})\big]v_z^2(\vec{q})\tau_z(\vec{q}) \cdot \Delta T\right\}\,, \tag{3}$$

whence one obtains the thermoconductivity

$$K = \frac{1}{(2\pi)^3}\int \mathrm{d}\vec{q}\frac{\partial}{\partial T}\big[\omega(\vec{q})n(\vec{q})\big] \cdot v_z^2(\vec{q})\tau_z(\vec{q})\,. \tag{4}$$

In the case of a uniform transport $v_z(\vec{q}) = v^2/3$, $\tau_z(\vec{q}) = \tau$, one obtains the classical result $K = Cq\Lambda/3$, where C is the specific heat, v^2 is the mean square velocity and $\Lambda = v\tau$ is the relaxation length (or mean free path). For a classical gas of particles Λ is the average inter-particle spacing and, as such, it is a constant as long as the number of particles is fixed. This is no longer true in the case of lattice vibrations where the number of phonons is not fixed and the heat transfer mechanism is different.

In the low-temperature limit the energy transfer is controlled by the collisions against the boundary walls. In this case it is well-known[2)] that the (non–thermal) relaxation time is given by

$$\sigma_z^0(\vec{q}) = \frac{d}{v_\perp(\vec{q})}\,, \tag{5}$$

where $v_\perp(\vec{q})$ is the group velocity along the transverse directions and d is of the order of the transverse size of the rod (shape-effects included). At non-vanishing temperatures the absorption probability is increased by the $n(\vec{q})$ factor, according to the radiation-like description of the phonons. Therefore, the (thermal) relaxation time is reduced to

$$\tau_z^{\mathrm{th}}(\vec{q}) = \frac{d}{v_\perp(\vec{q})n(\vec{q})} \,. \tag{6}$$

The usual Matthiessen rule[5)]

$$\frac{1}{\tau_z(\vec{q})} = \frac{1}{\tau_z^0(\vec{q})} + \frac{1}{\tau_z^{\mathrm{th}}(\vec{q})} \,, \tag{7}$$

may be used now, which interpolates between the thermal and non-thermal regime, with the result

$$\tau_z(\vec{q}) = \frac{d}{v_\perp(\vec{q})}\left[1 + n(\vec{q})\right]^{-1}. \tag{8}$$

The thermoconductivity (4) can now be written as

$$K = \frac{d}{(2\pi)^3 T^2}\int \mathrm{d}\vec{q}\frac{\omega^2(\vec{q})}{\exp\left[\omega(\vec{q})/T\right] - 1}\cdot\frac{v_z(\vec{q})}{v_\perp(\vec{q})} \,, \tag{9}$$

which, for a Debye spectrum to $\omega(\vec{q}) = vq$, becomes

$$\left\{\begin{aligned} K &= \frac{9\pi}{4} dnvQ(\omega_D/T) \,, \\ Q(x) &= \frac{1}{x^3}\int_0^x \mathrm{d}\xi\frac{\xi^4}{\mathrm{e}^\xi - 1} \,, \end{aligned}\right. \tag{10}$$

where $\omega_D = (6\pi^2 n)^{1/3} v$ is the Debye frequency and n is the number of unit cells per unit volume (a factor of 3 polarizations has also been included). The asymptotic behaviours of the thermoconductivity (10) are readily obtained as

$$\left\{\begin{aligned} K_{T\ll\omega_D} &\cong 54\pi dnv\left(\frac{T}{\omega_D}\right)^3 \simeq 0.73Cdv \,, \\ K_{T\gg\omega_D} &\cong \frac{9\pi}{16} dnv\frac{\omega_D}{T} \,. \end{aligned}\right. \tag{11}$$

where C is the specific heat. The reduced thermoconductivity Q given by (10) is plotted versus T/ω_D in Fig. 1, where one can see the peak located at $T \doteq 0.47\omega_D$. It is noteworthy that the high-temperature $1/T$–behaviour in (11) arises also from the low-frequency phonons, which control the relaxation length in this high-temperature region by their excessive thermal activation. Therefore, one may say that not only the T^3–law in the low-temperature limit but also the $1/T$–law in the high-temperature limit is a consequence of the quantum nature of the heat

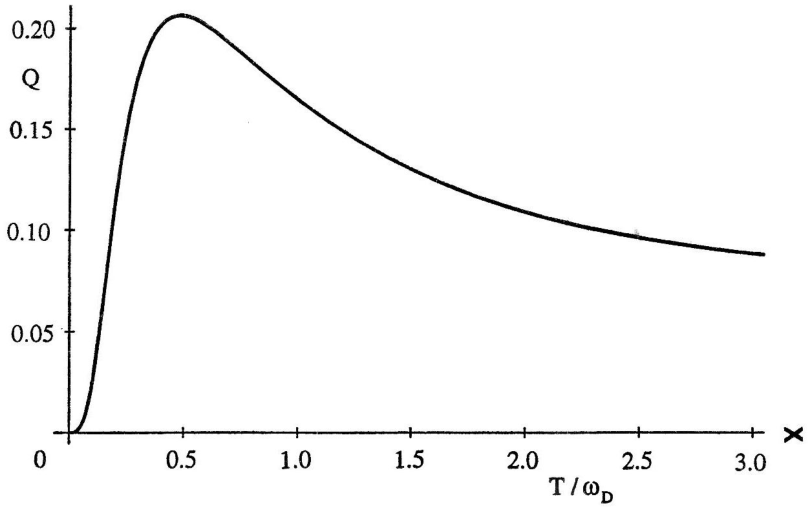

Fig. 1. The reduced thermoconductivity Q given by Eq. (10) versus the reduced temperature T/ω_D.

carriers. One may also add that for an anisotropic Debye spectrum $\omega(\vec{q}) = (v_\parallel^2 q_z^2 + v_\perp^2 q_\perp^2)^{1/2}$ the factor v in (10) is replaced by the ratio $v_\parallel^2/v_\perp$. In general, Eq. (9) can be used for any phonon branch, anisotropy included[6)].

In the case of non-equilibrium transport of energy (but preserving the local thermal equilibrium) one can derive, in the same manner that led to Eq. (4), the thermoconductivity

$$\bar{K} = \frac{d}{(2\pi)^3} \frac{\partial}{\partial T} \int \mathrm{d}\vec{q}\, \omega(\vec{q}) \cdot \frac{v_z^2(\vec{q})}{v_\perp(\vec{q})} \, \frac{n(\vec{q})}{1 + n(\vec{q})} \,, \tag{12}$$

related to the energy flow. It coincides with K in the low-temperature limit but behaves like

$$\bar{K} = \frac{9\pi}{20} dnv(\omega_D/T)^2 \tag{13}$$

in the high-temperature limit, where the energy transport is limited by the dissipation processes.

In conclusion, one may say that the lattice thermoconductivity of a rod of an ideal crystal has been derived without making any particular assumption on the specific mechanisms of heat transfer, except that the phonons are absorbed at the rod boundaries at a rate which is proportional to their occupation number. This is the natural assumption as long as the radiation-like description of the ideal phonons holds. However, various heat transfer mechanisms can be included in this picture, their effect being that of further reducing the relaxation time given by Eq. (8). For example, one may expect such a reduction to arise from the electron-phonon interaction. However, the number of electron states available for interaction is much smaller than the number of phonon states, the usual estimation being the ratio of the sound velocity to the Fermi velocity. Therefore, one may say that the lattice thermoconductivity is not much affected by the electron-phonon interaction. The presence of the impurities may also be included in Eq. (8). In particular, the well-known infrared divergencies[1] coming from the scattering length of the impurities are, according to (8), screened out, such that the thermoconductivity peak will be more or less affected, but not the asymptotic behaviours. The effect of the phonon-phonon interaction and of the crystal anharmonicities may also be taken into account. As long as these processes involve higher frequencies (as for the umklapp-scattering) they will mainly affect the peak of the thermoconductivity, decreasing the magnitude and shifting its position toward lower temperatures. Only where the low-frequency part of the Debye spectrum is damaged one may expect a departure from the ideal asymptotic laws (11). All these effects can most conveniently be accounted for by means of various techniques[7–11], among which the well-known variational approach to the Boltzmann equation. An interesting point would be to check Eqs. (10) and (11) against the available experimental data. This can be done most conveniently for compounds with low Debye frequency ω_D. Among the quasi-one-dimensional compounds one can often find the best candidates. A very interesting problem occurs, however, in this case, regarding the phason thermoconductivity (and, generally, the thermoconductivity of a lattice soft-mode). Equation (4) suits very well the anisotropy of these compounds, such that the computations might successfully be carried out. All these matters are left for forthcoming investigations.

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O TOPLINSKOJ VODLJIVOSTI IDEALNOG KRISTALA

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Opisujući nosioce topline (Debyeve fonone) analogno ponašanju fotona dobivena je temperaturna ovisnost toplinske vodljivosti rešetke u idealnom kristalu.