THEORY OF THE THERMAL CONDUCTIVITY OF THIN DIELECTRIC SAMPLES

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A theory of the thermal conductivity of thin dielectric plates is developed, taking into account the fact that the scattering of phonons by the boundaries of a sample becomes nearly specular when the temperature is reduced. The results depend strongly on the relationship between the characteristic dimensions of the roughness of boundary, the thickness of the sample, and the bulk parameters of the dielectric.

IT is known that the principal reason for the thermal resistance of thin samples at sufficiently low temperatures is the scattering of phonons at crystal boundaries. It is usually assumed that the scattering of phonons is diffuse. Because the phonons lose their ordered motion in the diffuse scattering process, the effective mean free path of the phonons l^{eff} is of the order of the thickness of the sample d. Since the order of magnitude of the thermal conductivity is $\kappa \approx l^{\text{eff}} sC$ (s is the velocity of sound and C is the specific heat), and since at low temperatures $C \propto T^3$, we find that $\kappa \propto dT^3$ in the diffuse scattering case (cf. [1]).

Such a conclusion is justified when the average phonon wavelength λ_T is much shorter than the characteristic dimensions of the surface roughness η. However, $\lambda_T \approx \hbar s/T$ ($\hbar\omega = sh/\lambda_T \approx T$) and, therefore, when the temperature is lowered a boundary becomes smoother in relation to the scattering phonons and, consequently, l^{eff} should increase when the temperature is lowered.[1)] We shall show here that allowance for this change alters considerably the temperature dependence of the thermal conductivity. The final result depends on the relationship between η, d, and the bulk parameters of the dielectric (cf. Figs. 1 and 2).

We shall now analyze the problem quantitatively. We shall consider the thermal conductivity of a single-crystal plate of thickness d, which is small compared with the mean free path l for the scattering in the interior: $d \ll l$. In practice, this inequality may be satisfied at low temperatures, when the frequency of collisions between phonons, accompanied by umklapp processes, is small compared with the frequency of normal collisions. Therefore, the mean free path l is related either to normal collisions or to the scattering of phonons by microscopic lattice defects. We shall also assume that the phonon dispersion law is linear and isotropic: $\hbar\omega = sq$ ($\mathbf{q}$ is the phonon momentum).

The mechanism of interaction of phonons with the boundary of a sample is important for any further analysis. This problem has been considered by Ziman [2] when the phonons are incident normally on a boundary, on the assumption that the roughness of the boundary is distributed in accordance with the Gaussian law.

Ziman's treatment can be generalized without difficulty to an arbitrary angle of incidence ϑ, and gives the following result for the probability P of specular reflection of phonons:

$$P = \exp(-4\pi^2\eta^2\lambda^{-2}\cos^2\vartheta), \tag{1}$$

where the angle ϑ is measured from the normal to the surface.

If we select the z axis to be perpendicular to the surface of a film and the x axis to lie along a temperature gradient, the linearized transport equation for the distribution function $n = n_0 + g[n_0(\omega, x)$ is the equilibrium value of the distribution function at a given temperature and

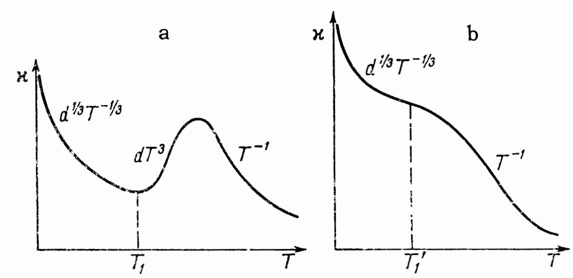

FIG. 1.

[1)]Heat is transported in a thin plate by "glancing" phonons, traveling almost parallel to the boundary. The scattering of these phonons becomes nearly specular when $\lambda_T \ll \eta$.

$g(\mathbf{q}, z)$ is a small correction] will have the form

$$v_z \frac{\partial g}{\partial z} + v_x \nabla T \frac{\partial n_0}{\partial T} = \hat{J} g, \qquad (2)$$

where $\hat{J}$ is the collision operator. Equation (2) should be supplemented by the boundary conditions (a P-th fraction of phonons is reflected specularly from the boundary):

$$g(\mathbf{q}, d) = P g(\mathbf{q}', d), \quad q_z < 0,$$
$$g(\mathbf{q}, -d) = P g(\mathbf{q}', -d), \quad q_z > 0, \qquad (3)$$

where $\mathbf{q}' = (q_x, q_y, -q_z)$, and 2d is the thickness of the plate.

Equation (2) is a complex integro-differential equation, which cannot be solved in its general form. However, it is known that transport processes in thin films (as in the anomalous skin effect) are governed mainly by the action of the so-called "glancing" quasi-particles incident almost parallel to the surface.[3,4] The quantity g is a critical function of the angle ϑ, and we can introduce a relaxation time $\hat{J} g = -g/\tau$. However, in the present problem, the relaxation time approximation is not always justified; we shall consider this point again. After this substitution, Eq. (2) simplifies:

$$v_z \frac{\partial g}{\partial z} + \frac{g}{\tau} = -v_x \nabla T \frac{\partial n_0}{\partial T}. \qquad (4)$$

Bearing in mind that the thermal flux is

$$U_x = \frac{3}{h^3} \int \hbar \omega v_x g \, d\mathbf{q},$$

we obtain, by solving Eq. (4) with the boundary conditions (3) and subsequent averaging of U_x over z:

$$\bar{U}_x = \nabla T \frac{2\pi T^3}{h^3 s^2} \int_0^\infty dw \int_0^1 du \, (1-u^2) \, l$$
$$\times \left[1 - \frac{u}{k} \frac{(1-e^{-\alpha u^2})(1-e^{-k/u})}{1-\exp(-\alpha u^2 - k/u)} \right] \frac{w^4 e^w}{(e^w - 1)^2}. \qquad (5)$$

In this formula, the following notation is used

$$\omega = \hbar\omega/T, \quad u = \cos\theta, \quad k = 2d/l, \quad \alpha = 4\pi^2\eta^2/\lambda^2. \qquad (6)$$

Using the fact that α and k are power functions of w, we can easily show that, when the integration is carried out with respect to w in Eq. (5), the main contribution is made by $w \approx 1$. From now on, we shall understand α and k to be the "temperature" values of these quantities, taken at $w \approx 1$. In this approximation, the result (5) may be written in the form

$$\varkappa = {}^1/_3 C s l^{\text{eff}}. \qquad (7)$$

Here

$$C = {}^{12}/_5 \pi^4 N (T/\Theta)^3,$$
$$l^{\text{eff}} = \frac{3}{2} l \int_0^1 du \, (1-u^2) \left[1 - \frac{u}{k} \frac{(1-e^{-\alpha u^2})(1-e^{-k/u})}{1-e^{-\alpha u^2 - k/u}} \right], \qquad (8)$$

where N is the number of atoms per unit volume and Θ is the Debye temperature.

We have shown that the relaxation time approximation is justified if g is a critical function of the angle ϑ. Using the explicit expression for g, we can easily see that for this approximation to apply we must have $k \ll \alpha$, and consequently Eq. (8) is valid only when this inequality is satisfied.

Since the integral cannot be used in its general form, we shall consider the most interesting limiting cases, assuming that $k \ll \alpha$ (and obviously $k \ll 1$).

When $\alpha k^2 \gg 1$, we find after simple calculations

$$l^{\text{eff}} \cong {}^3/_2 d \ln(l/2d). \qquad (9)$$

This agrees (to within an unimportant logarithmic factor) with the results of Casimir,[1] which is as expected because $u \approx k$ for the glancing phonons and, according to Eq. (1), the probability of specular reflection $P \approx \exp(-\alpha k^2)$ tends to zero for such phonons.

In the other limiting case, $\alpha k^2 \ll 1$, when the integral in Eq. (8) is being calculated the integration range can be conveniently split into three intervals, in the first of which $\alpha u^2 \ll 1$, in the second $\alpha u^2 + k/u \ll 1$, and in the third $k/u \ll 1$ (the conditions $\alpha k^2 \ll 1$ and $k \ll \alpha$ ensure that these intervals overlap). The result has the form

$$l^{\text{eff}} \cong \frac{\pi}{3\sqrt{3}} l \left(\frac{k}{\alpha} \right)^{1/3}. \qquad (10)$$

The results obtained above are valid for $k \ll \alpha$. In thin samples, we can also have the case $\alpha \ll k \ll 1$. It follows from this inequality that the mean free path of any phonon scattered diffusely by the boundaries in a film is much greater than l:

$$\frac{d}{1-P} \approx \frac{d}{\alpha u^2} \gg l. \qquad (11)$$

In other words, a phonon loses its glancing nature earlier if it suffers diffuse scattering. Clearly, under such conditions, the glancing phonons cannot play a distinct role and, therefore, we cannot introduce a relaxation time and cannot solve the problem exactly. However, qualitatively correct results can be obtained from the following simple considerations. (It should be remembered that normal collisions conserve momentum and, therefore, they of themselves do not give rise to a finite thermal conductivity.)

If the scattering by local defects predominates, then, as clearly follows from Eq. (11), the influence of the boundaries can be neglected and the sample can be regarded as bulky up to $\alpha \sim k$:

$$l^{\text{eff}} \approx l. \qquad (12)$$

As expected, this result follows from Eq. (8).

In sufficiently pure samples, the normal collisions predominate and their role reduces to making the phonon distribution isotropic. A more detailed analysis shows that in this case phonons traveling along all directions make approximately the same contribution and, therefore,

$$l^{\text{eff}} \approx d/\alpha. \qquad (13)$$

Let us now gather together all the results obtained so far. We recall that, when $T \ll \Theta$, the mean free path in normal collisions is $l^N \approx l_\Theta (\Theta/T)^5$ and in collisions with local defects it is $l_i \approx l_0(\Theta/T)^4$; the orders of magnitude are $l_\Theta \approx aMs^2/\Theta$, $l_0 \approx a/c$, where M is the mass of an atom, a is the lattice constant, and c is the concentration of defects. We should mention also that, according to Eq. (6), $\alpha \approx (\eta T/a\Theta)^2$ and $k \approx d/l$.

First, we shall consider first relatively high temperatures, for which $l(T) \lesssim d$.

1. We shall begin with "impure" samples in which scattering by defects predominates; more exactly, when $l_i \sim d$, $l^N \gg d$. There are two possible cases.

A. If $\alpha \gtrsim 1$ when $l_i \sim d$, then, according to Eqs. (7), (9), and (10),

$$\varkappa \sim \begin{cases} dT^3, & \alpha k^2 \gg 1 \\ d^{1/3}T^{-1/3}, & \alpha k^2 \ll 1 \end{cases}.$$

These dependences are shown in Fig. 1a. The law $\kappa \propto 1/T$ corresponds to a bulk sample. The temperature $T_1 \approx \Theta(al_0/\eta d)^{1/5}$ is given by the condition $\alpha k^2 \approx 1$.

B. If $\alpha \ll 1$ when $l_i \sim d$, then, according to Eqs. (12) and (10),

$$\varkappa \sim \begin{cases} T^{-1}, & \alpha \ll k \\ d^{1/3}T^{-1/3}, & \alpha \gg k \end{cases}.$$

In Fig. 1b the temperature $T_1' \approx \Theta(\eta l_0/ad)^{1/2}$ is found from the condition $\alpha \sim k$.

2. In the case of relatively pure samples (when $l^N \sim d$ and $l_i \gg d$), we also have two possible cases.

A. If $\alpha \gtrsim 1$ when $l^N \approx d$, then, according to Eqs. (9) and (10),

$$\varkappa \sim \begin{cases} dT^3, & \alpha k^2 \gg 1 \\ d^{1/3}T^{-1}, & \alpha k^2 \ll 1 \end{cases}.$$

The exponential rise of $\kappa(T)$, shown in Fig. 2a, is due to umklapp processes under those conditions when the sample can be regarded as bulky. The dependence $\kappa \propto d^2T^8$ is associated with the hydrodynamic mechanism of thermal conductivity, which applies when $d \gg l^N \gg d^2/l^U$, where l^U is the mean free path for phonon-phonon collisions accompanied by umklapp, $l^U \gg l^N$ (cf. a paper of one of the present authors[5]). The temperature is

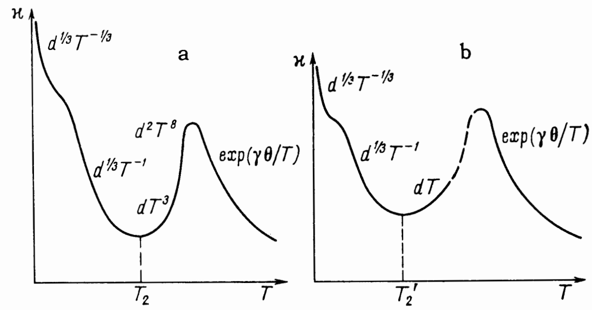

FIG. 2.

$T_2 \approx \Theta(al_\Theta/\eta d)^{1/3}$. When the temperature is reduced, collisions with defects become more likely than normal collisions. In plotting the curve, it has been assumed that this occurs only in the region $\alpha k^2 \ll 1$.

B. If $\alpha \ll 1$ when $l^N \sim d$, then, according to Eqs. (13) and (10),

$$\varkappa \sim \begin{cases} dT, & \alpha \ll k \\ d^{1/3}T^{-1}, & \alpha \gg k \end{cases}.$$

Figure 2b shows dashed the range in which $l^N \ll d \ll l^U$ and $\alpha \ll 1$. This case requires special treatment, which has not yet been carried out. The temperature is $T_2' \approx \Theta(\eta l_\Theta/ad)^{1/3}$.

Under experimental conditions, cases 1A and 2A are easier to realize. For example, case 2A is realized when $d \approx 10^{-2}$ cm, $\eta/a \approx 10^2$ and $c \lesssim 10^{-6}$; the temperature is $T_2 \approx \Theta/10$. Cases 1B and 2B are possible only in bulky plates with very smooth surfaces (for a typical dielectric, these cases require $\eta/a \lesssim 10$ and $d \gtrsim 0.1$ cm).

In conclusion, we note that the main results obtained are not directly associated with the expression (1) for the probability of specular reflection P. In fact, it is evident from general considerations that P is an even function of the parameter $z = \eta u/\lambda$, where $P(z \ll 1) \approx 1 - \gamma z^2$ and $P(z \gg 1) \to 0$, $\gamma > 0$.

[1] H. B. G. Casimir, Physica 5, 495 (1938).

[2] J. M. Ziman, Electrons and Phonons, Cambridge University Press, 1960 (Russ. Transl., IIL, 1962).

[3] M. Ya. Azbel' and É. A. Kaner, JETP 32, 896 (1957), Soviet Phys. JETP 5, 730 (1957).

[4] M. Ya. Azbel' and R. N. Gurzhi, JETP 42, 1632 (1962), Soviet Phys. JETP 15, 1133 (1962).

[5] R. N. Gurzhi, JETP 46, 719 (1964), Soviet Phys. JETP 19, 490 (1964).

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