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Effect of Normal Processes on the Thermal Conductivity of Finite Insulating Samples.

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Summary. — A model for the calculation of the thermal conductivity of finite dielectric samples is given. Explicit calculations have been made for two different geometries in the two limiting cases where the phonon-phonon mean free path is much longer and much shorter respectively than the characteristic dimensions of the sample.

1. – Introduction.

The object of the present paper is to study the influence of the normal processes between phonons on the thermal conductivity of insulators at low temperatures.

Basically, we follow the kinetic method suggested by CHAMBERS (1) but, in order to take into account the fact that normal processes do not change the total momentum we assume that the collisions relax the system towards a displaced distribution function. We assume, also, that the density of particles scattered from the boundaries is given by the equilibrium distribution function at the local temperature (diffuse boundary scattering).

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(1) R. G. CHAMBERS: *Proc. Phys. Soc.*, A **65**, 458 (1952).

We consider the crystal to be free of imperfections and since we are interested in the region of low temperatures, we neglect umklapp processes.

Experiments carried out by WHITWORTH [2] in liquid helium demostrate that, similarly to what happens in a fluid, below a certain critical value of the mean free path l of the phonon-phonon interactions the energy flux increases monotonically as l decreases. In these experiments it is shown that the thermal conductivity has a minimum for l of the order of d, d being some characteristic dimension of the sample (Knudsen minimum effect).

According to Whitworth's experiments, normal collisions act in such a way that when l is much larger than d, the main resistive effect is due to boundary scattering. As l decreases, the normal collisions modify the directions of the phonons, therefore a larger number of them can be absorbed by the walls and consequently the heat flux is reduced. In the opposite limit, when l becomes sufficiently small, the phonons travelling inside the sample will not reach the walls as easily as they did for greater l. The ensuing situation resembles the flux of a viscous fluid under a pressure gradient in that an ordered motion is established. This ordered motion is characterized by a velocity field $\boldsymbol{\lambda}(\boldsymbol{r})$ which depends on the position inside the crystal.

GUYER and KRUMHANSL [3] have proposed an equation for the thermal conductivity of finite samples with both normal and resistive processes present. When the mean free path for resistive processes tends to infinity this equation would give the thermal conductivity of a perfect crystal. The formula thus obtained reproduces the results of SUSSMANN and THELLUNG [4] as well as those of GURZHI [5] and is in accord with our results in this paper in the case $l/d \ll 1$. In the opposite limit ($l = \infty$), Guyer and Krumhansl's formula reproduces Casimir's results. However, the correction to first order in d/l is linear, whereas both in the present paper and in those by KARCHAVA and SANIKIDZE [6], while they still reproduce Casimir's results in the case $l = \infty$ the first-order correction in d/l, is of the form $d/l \cdot \ln(d/l)$.
KARCHAVA and SANIKIDZE [6] have solved the Boltzmann equation by using the relaxation-time approximation for the case of weak normal scattering between phonons. These authors assume that the normal processes make the distribution function tend to the Bose-Einstein equilibrium distribution $f^0(\varepsilon_q)$ instead of a displaced distribution $f = f^0(\varepsilon_q - \hbar\boldsymbol{\lambda}\boldsymbol{q})$ as in this paper. In the above formulae ε_q and $\boldsymbol{q}$ are the energy and wave number of the phonons respectively. These assumptions prove to be equivalent for $l \gg d$ scattering.

([2]) R. W. WHITWORTH: *Proc. Roy. Soc.*, A **246**, 390 (1958).
([3]) R. A. GUYER and J. A. KRUMHANSL: *Phys. Rev.*, **148**, 778 (1966).
([4]) J. A. SUSSMANN and A. THELLUNG: *Proc. Phys. Soc.*, **81**, 1122 (1963).
([5]) R. N. GURZHI: *Sov. Phys. JETP*, **19**, 490 (1964).
([6]) T. A. KARCHAVA and D. G. SANIKIDZE: *Sov. Phys. JETP*, **23**, 1118 (1966).

In Sect. 2 we describe the model used to study the influence of the normal processes betwen phonons in finite dielectric samples. We obtain an integral equation for $\boldsymbol{\lambda}(\boldsymbol{r})$ which allows us to evaluate the thermal conductivity K for any geometry and scattering intensity.

In Sect. 3, we solve this integral equation for a slab and a cylinder in the limiting cases $l \gg d$ and $l \ll d$, d being the thickness of the slab and the diameter of the cylinder respectively. In both limits our results agree with those obtained in [5,6].

2. – Formulation of the model.

In this Section we propose a model for the heat conduction problem in finite insulator samples. It is further assumed that a thermal gradient is applied and that only normal processes can occur.

The kinetic method suggested by CHAMBERS has been successfully applied to solve transport problems in metals when size effects [7] have to be taken into account.

We apply here the same method to the problem of thermal conductivity in insulators. We propose then, the following form for the distribution function:

$$f(\boldsymbol{r}, \boldsymbol{q}) = g(\boldsymbol{r}_B, \boldsymbol{q}) \exp\left[-\frac{|\boldsymbol{r}-\boldsymbol{r}_B|}{l}\right] + \int_0^{s_0} f_\lambda(\boldsymbol{r}', \boldsymbol{q}) \exp\left[-\frac{s_0 - s}{l}\right]\frac{\mathrm{d}s}{l}. \tag{2.1}$$

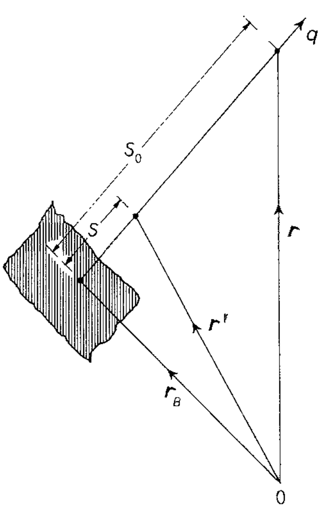

Fig. 1. – The vector $\boldsymbol{r}$, $\boldsymbol{q}$ and $\boldsymbol{r}_B$ used in the text are drawn in this Figure.

The number of particles with wave vector $\boldsymbol{q}$, emitted from the boundaries at a point $\boldsymbol{r}_B$ is given by $g(\boldsymbol{r}_B, \boldsymbol{q})$. Accordingly, the first term of (2.1) gives the number of carriers emitted from $\boldsymbol{r}_B$ and reaching $\boldsymbol{r}$ in direction $\boldsymbol{q}$. The second term in (2.1) represents the sum of the contributions of particles that, due to collisions enter the trajectory defined by $\boldsymbol{r}$ and $\boldsymbol{q}$ at $\boldsymbol{r}'$. (See Fig. 1).

In writing (2.1) we assume that the prob-

[7] R. G. CHAMBERS: *Proc. Roy. Soc.*, A **202**, 378 (1950).

ability for a particle to travel a distance s without collision is $\exp[-s/l]$ where l is the mean free path.

We also assume that the number of particles entering an element of trajectory $\mathrm{d}s$ at $\boldsymbol{r}$ with a direction $\boldsymbol{q}$ is $f_\lambda(\boldsymbol{r}, \boldsymbol{q})\cdot \mathrm{d}s/l$. The explicit form of $f_\lambda(\boldsymbol{r}, \boldsymbol{q})$ has been given in Sect. **1**.

It can be shown by derivation that, for $g(\boldsymbol{r}_B, \boldsymbol{q})$ and $f_\lambda(\boldsymbol{r}, \boldsymbol{q})$ known, the distribution function given by eq. (2.1) satisfies the following Boltzmann equation:

$$\boldsymbol{c}\nabla_r f(\boldsymbol{r}, \boldsymbol{q}) = \frac{f_\lambda(\boldsymbol{r}, \boldsymbol{q}) - f(\boldsymbol{r}, \boldsymbol{q})}{\tau}, \tag{2.2}$$

where $\tau = l/c$ and $\boldsymbol{c}$ is the phonon velocity.

The drift velocity λ is to be determined in a way similar to that followed by CALLAWAY (8) by imposing local momentum conservation:

$$\int \frac{f_\lambda(\boldsymbol{r}, \boldsymbol{q}) - f(\boldsymbol{r}, \boldsymbol{q})}{\tau}\, \boldsymbol{q}\, \mathrm{d}^3 q = 0\,. \tag{2.3}$$

If the scattering at the walls is diffuse $g(\boldsymbol{r}_B, \boldsymbol{q})$ has to be taken equal to $f^0(\boldsymbol{r}_B, \boldsymbol{q})$, the local equilibrium Bose-Einstein distribution.

By partial integration in (2.1) we obtain

$$f(\boldsymbol{r}, \boldsymbol{q}) = f^0(\boldsymbol{r}_B, \boldsymbol{q})\exp[-s_0/l] + f_\lambda(\boldsymbol{r}, \boldsymbol{q}) - f_\lambda(\boldsymbol{r}_B, \boldsymbol{q})\exp[-s_0/l] - \\ - \exp[-s_0/l]\int_0^{s_0} \exp[s/l]\frac{\mathrm{d}f_\lambda}{\mathrm{d}s}\,\mathrm{d}s\,; \tag{2.4}$$

for $\lambda/c \ll 1$, we can approximate

$$f_\lambda \simeq f_0 + \frac{\hbar\boldsymbol{\lambda}\cdot\boldsymbol{q}}{kT} f^0(f^0+1)\,, \tag{2.5}$$

where k is the Boltzmann constant and T the absolute temperature.

And therefore (2.4) reduces to

$$f(\boldsymbol{r}, \boldsymbol{q}) - f_\lambda(\boldsymbol{r}, \boldsymbol{q}) = -\frac{\hbar}{kT}\boldsymbol{\lambda}(\boldsymbol{r}_B)\cdot \boldsymbol{q} f^0(f^0+1)\exp[-s_0/l] - \\ - \int_0^{s_0} \exp\left[-\frac{s_0 - s}{l}\right]\frac{\mathrm{d}f_\lambda}{\mathrm{d}s}\,\mathrm{d}s\,. \tag{2.6}$$

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(8) J. CALLAWAY: *Phys. Rev.*, **113**, 1046 (1959).

On the other hand,

$$\frac{\mathrm{d}f_\lambda}{\mathrm{d}s}=\frac{\mathrm{d}f_\lambda}{\mathrm{d}\lambda}\frac{\mathrm{d}\lambda}{\mathrm{d}s}+\frac{\mathrm{d}f_\lambda}{\mathrm{d}T}\nabla T\cdot\frac{\boldsymbol{c}}{c}\,. \tag{2.7}$$

According to (2.7) we have for (2.6)

$$\begin{aligned} f(\boldsymbol{r})-f_\lambda(\boldsymbol{r}) &= -\frac{\hbar}{kT}\lambda(\boldsymbol{r}_B)\,q_z f^0(f^0+1)\exp[-s_0/l]-\\ &\quad -l\,[1-\exp[-s_0/l]]\frac{\hbar q_z}{kT}\frac{\mathrm{d}T}{\mathrm{d}z}\frac{c}{T}f^0(f^0+1)-\\ &\quad -\frac{\hbar q_z}{kT}f^0(f^0+1)\int_0^{s_0}\exp\left[-\frac{s_0-s}{l}\right]\frac{\mathrm{d}\lambda}{\mathrm{d}s}\,\mathrm{d}s\,, \end{aligned} \tag{2.8}$$

for brevity we omit the explicit dependence of the functions on $\boldsymbol{q}$. q_z is the z-component of the wave vector $\boldsymbol{q}$. Integrating by parts the last term of (2.8) we obtain

$$\begin{aligned} f(\boldsymbol{r})-f_\lambda(\boldsymbol{r}) &=\\ &= -\frac{\hbar q_z}{kT}f^0(f^0+1)\lambda(r)-l\,[1-\exp[-s_0/l]]\frac{\hbar q_z}{kT}f^0(f^0+1)\frac{c}{T}\frac{\mathrm{d}T}{\mathrm{d}z}+\\ &\quad +\frac{\hbar q_z}{kT}f^0(f^0+1)\frac{1}{l}\int_0^{s_0}\lambda(s)\exp\left[-\frac{s_0-s}{l}\right]\mathrm{d}s\,. \end{aligned} \tag{2.9}$$

Then, the local-momentnm-conservation condition (2.3) has the form

$$\begin{aligned} &\lambda(\boldsymbol{r})\int\mathrm{d}^3q\,q_z^2 f^0(f^0+1)-\frac{1}{l}\int\mathrm{d}^3q\left[\int_0^{s_0}\lambda(s)\exp\left[-\frac{s_0-s}{l}\right]\mathrm{d}s\right]q_z^2 f^0(f^0+1)=\\ &\quad=-\frac{c}{T}l\frac{\mathrm{d}T}{\mathrm{d}z}\left[\int\mathrm{d}^3q\,q_z^2 f^0(f^0+1)-\int\mathrm{d}^3q\,q_z^2 f^0(f^0+1)\exp[-s_0/l]\right]. \end{aligned} \tag{2.10}$$

This equation determines $\lambda(\boldsymbol{r})$ completely.

Notice also from (2.3) that, in the case in which τ is independent of wave vector, the heat current density $\overline{\boldsymbol{U}}$ can be calculated directly from $f_\lambda(\boldsymbol{r},\boldsymbol{q})$:

$$\overline{\boldsymbol{U}}=\frac{1}{S_c}\iint\mathrm{d}S_c\,\mathrm{d}^3q\,f_\lambda(\boldsymbol{r},\boldsymbol{q})\,\varepsilon_q\,\boldsymbol{c}\,, \tag{2.11}$$

where S_c is the area of a section of the sample normal to the direction of heat flow.

3. – Application of the model.

In this Section we find solutions of (2.10) for two particular geometries, a slab of thickness d and a cylinder of radius $R = d/2$ and for the limiting cases: $l \gg d$ and $l \ll d$. We assume that l does not depend on the wave number q.

3·1. *Case of the slab.* – We want now to particularize (2.10) for the case of the slab of thickness d. A small temperature gradient is applied along the z-axis. The y-axis is taken perpendicular to the walls defined by the $y = 0$ and $y = d$ planes (see Fig. 2).

For this geometry, (2.10) must be written in the form

$$(3.1)\qquad \frac{4\pi}{3}\lambda(y) - \frac{1}{l}\int_0^{2\pi}\mathrm{d}\varphi\left[\int_0^{\pi/2}\mathrm{d}\theta\sin^3\theta\cos^2\varphi\int_0^y\exp\left[-\frac{y-y'}{l\cos\theta}\right]\lambda(y')\frac{\mathrm{d}y'}{\cos\theta} + \right.$$

$$\left. + \int_{\pi/2}^{\pi}\mathrm{d}\theta\sin^3\theta\cos^2\varphi\int_d^y\exp\left[\frac{y-y'}{l|\cos\theta|}\right]\lambda(y')\frac{\mathrm{d}y'}{\cos\theta}\right] =$$

$$= -\frac{c}{T}\frac{\mathrm{d}T}{\mathrm{d}z}l\frac{4\pi}{3} + \frac{c}{T}\frac{\mathrm{d}T}{\mathrm{d}z}l\int_0^{2\pi}\mathrm{d}\varphi\left[\int_0^{\pi/2}\sin^3\theta\cos^2\varphi\exp\left[-\frac{y}{l\cos\theta}\right]\mathrm{d}\theta + \right.$$

$$\left. + \int_{\pi/2}^{\pi}\sin^3\theta\cos^2\varphi\exp\left[-\frac{d-y}{l|\cos\theta|}\right]\mathrm{d}\theta\right].$$

For $l \gg d$, we can neglect the second term on the left-hand side. Then, we obtain

$$(3.2)\qquad \lambda(y)|_{l\gg d} = = \frac{3}{4}\frac{c}{T}\frac{\mathrm{d}T}{\mathrm{d}z}\left[y\ln\frac{y}{l} + (d-y)\ln\frac{d-y}{l}\right].$$

It can be easily seen that the term we neglected is of the order of

$$\frac{d}{l}\left(\ln\frac{d}{l}\right)^2.$$

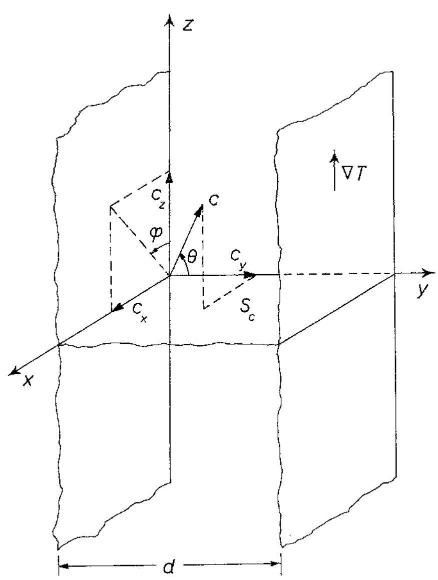

Fig. 2. – Slab geometry. The co-ordinates used in the text are shown.

For $l \ll d$ we neglect the second term of the right-hand side. Accordingly, we have the following integral equation to solve:

$$(3.3)\qquad \lambda(y) - \frac{3}{4l}\left[\int_0^{\pi/2} d\theta \sin^3\theta \int_0^y \exp\left[-\frac{y-y'}{l\cos\theta}\right]\lambda(y')\frac{dy'}{\cos\theta} + \int_{\pi/2}^{\pi} \sin^3\theta\, d\theta \int_d^y \exp\left[\frac{y-y'}{l|\cos\theta|}\right]\lambda(y')\frac{dy'}{\cos\theta} = -\frac{c}{T}\frac{dT}{dz}l.$$

The solution of (3.3) for all points farther than 1 from the boundaries is

$$(3.4)\qquad \lambda(y)|_{l\ll d} = -\frac{5}{2}\frac{c}{T}\frac{dT}{dz}\frac{1}{l}y(d-y).$$

As expected in this limit $\lambda(y)$ increases as l decreases. This corresponds to a Poiseuille flow of phonons.

3·2. *Case of the cylinder.* – We consider here a circular cylinder of diameter d with a small uniform temperature gradient applied along its axis (see Fig. 3).

Equation (2.10) takes the form

$$(3.5)\qquad \lambda(\varrho) - \frac{3}{4\pi l}\int_0^{2\pi} d\varphi \int_0^{\pi} d\theta \sin\theta \cos^2\theta \int_0^{t_0} \lambda(t)\exp\left[-\frac{t_0-t}{l\sin\theta}\right]\frac{dt}{\sin\theta} = -\frac{c}{T}\frac{dT}{dz}l + \frac{c}{T}\frac{dT}{dz}l\frac{3}{4\pi}\int_0^{2\pi} d\varphi \int_0^{\pi} d\theta \sin\theta\cos^2\theta \exp\left[-\frac{t_0}{l\sin\theta}\right],$$

where

$$t_0 = \varrho\cos\varphi + \sqrt{R^2 - \varrho^2\sin^2\varphi}$$

and R is the radius of the cylinder.

For $l \gg R$ we can neglect, as in the case of the slab, the second term of the l.h.s. in eq. (3.5). Then, we obtain the following expression for the drift velocity:

$$(3.6)\qquad \lambda(\varrho)|_{l\gg R} = -\frac{3}{2}\frac{c}{T}\frac{dT}{dz}R\left[E\left(\frac{\varrho}{R}, \frac{\pi}{2}\right) + f\left(\frac{\varrho}{l}\right)\right],$$

where $E(\varrho/R, \pi/2)$ is the elliptic integral of the

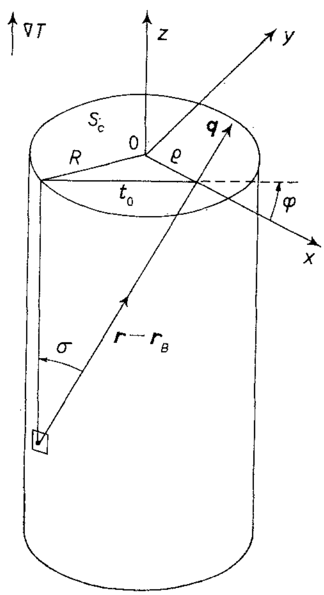

Fig. 3. – Cylindrical geometry. The co-ordinates used in the text are shown.

second kind [9] and the contribution of the leading term in $f(\varrho/l)$ to the conductivity is of the form $(R/l)\ln(R/l)$.

Formulae (3.2) and (3.6) agree with those obtained in ref. [6].

Notice that when l goes to infinity, we obtain from (3.6) the well-known Casimir's formula [10] for the thermal conductivity of a cylinder:

$$K = \tfrac{1}{3} C_v c d \, .$$

It is interesting to note also that for $l \gg d$ the drift velowity decreases with l.

For $l \ll d$ the solution of (3.5) is similar to that obtained for the slab in the same limit:

$$\lambda(\varrho)|_{l\ll R} = -\frac{5}{4}\frac{c}{T}\frac{\mathrm{d}T}{\mathrm{d}z}\frac{1}{l}(R^2-\varrho^2)\, . \tag{3.7}$$

This again corresponds to a Poiseuille flow of particles and is in agreement with the results obtained in ref. [3-5].

The results shown in this Section agree qualitatively with the results of the experiments made by WHITWORTH with cylinders.

4. – Discussion.

A formula has been obtained for the thermal conductivity of finite insulating samples, assuming that the effect of resistive processes (both Umklapp and those due to imperfections) may be neglected. In order that these results may be observed experimentally in solids it is necessary to have available crystals that are sufficiently devoid of imperfections.

In the temperature region where Poiseuille type flow is established this condition is given by (ref. [3,5])

$$\frac{l_N}{d} \ll 1 \qquad \text{and} \qquad \frac{d^2}{l_N} \ll l_R\, ,$$

where d is the diameter of the sample and l_N and l_R are respectively the mean free paths for normal and resistive processes.

As was mentioned on comparing our results with those of ref. [3], in the temperature region where ballistic flow is established ($l/d \gg 1$) normal processes are just as effective in hindering the flow of heat as resistive ones; therefore in this region the necessary condition to be able to neglect the effect of the latter is

$$\frac{l_N}{l_R} \ll 1\, .$$

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[9] E. JAHNKE and F. EMDE: *Tables of Functions with Formulae and Curves*, 4th Edition (New York, 1964), p. 54.

[10] H. B. G. CASIMIR: *Physica*, **5**, 495 (1938).

Let us now suppose that the conditions

$$l_R \gg \frac{d^2}{N} \qquad \text{and} \qquad \frac{d}{l_N} \gg 1$$

are satisfied at a given temperature T_1. Since l_R increases with decreasing temperature T (at worst, when l_R is solely due to macroscopic imperfections it remains constant), whereas l_N increases as T is lowered (approximately as l/T^5) it is clear that by decreasing T it is possible to reach a region where the conditions $d \ll l_N \ll l_R$ hold.

Since Poiseuille-type flow has been observed in the experiments in ref. ([11]), we conclude that in those same crystals one should be able to observe the effect of normal processes under the conditions in which ballistic flow is established.

In conclusion, the conditions under which resistive processes may be neglected in the regions of both ballistic and Poiseuille flow have become established. The authors have not been able to derive the corresponding condition in the intermediate region.

([11]) L. P. MEZHOV-DEGLIN: *Sov. Phys. JETP*, **22**, 47 (1966).

RIASSUNTO (*)

Si fornisce un modello per calcolare la conduttività termica dei campioni dielettrici finiti. Si eseguono calcoli espliciti per due differenti geometrie nei due casi limite dove il cammino libero medio fonone-fonone è rispettivamente molto più lungo e molto più corto delle dimensioni caratteristiche del campione.

(*) *Traduzione a cura della Redazione.*

Влияние нормальных процессов па термопроводимость конечных изоляционных образцов.

Резюме (*). — Предлагается модель для вычисления термопроводимости конечных диэлектрических образцов. Проведены точные вычисления для двух различных геометрий, в двух предельиых случаях, когда средняя фонон-фононная свободная длина, соответственно, много больше н много меньше, чем характеристические размеры образца.

(*) *Переведено редакцией.*