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THERMAL CONDUCTIVITY OF DIELECTRICS AND FERRODIELECTRICS AT LOW TEMPERATURES

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The thermal conductivity of dielectrics is studied in the low temperature region when collisions between quasiparticles (phonons or spin waves) accompanied by Umklapp processes are much less probable than collisions without Umklapps. It is shown that these normal collisions, which by themselves do not lead to thermal resistance, can significantly impede the transfer of momentum from the gas of quasiparticles to the boundaries of the sample or to other macroscopic distortions (for example, to dislocations or to the faces of single crystals) in sufficiently large and pure samples. Taking this fact into account leads to a significant change both in the magnitude of the coefficient of thermal conductivity and in its dependence on temperature and on the dimensions of the sample.

As is well-known, thermal resistance appears in an ideal single crystal of infinite dimensions only because of those collisions between quasiparticles which are accompanied by Umklapp processes. In the region of low temperatures the corresponding effective mean free path l^U increases exponentially with decrease of temperature: $l^U \sim e^{\gamma\Theta/T}$, where Θ is the Debye temperature if the problem involves phonons or the Curie temperature for the case of spin waves, T is the temperature, and $\gamma \sim 1$. It is clear that with a lowering of the temperature in a sufficiently pure sample, collisions of the quasiparticles with the boundaries begin to play a dominant role, and the effective mean free path l^{eff} turns out to be the order of the transverse dimension d of the sample.[1]

For a rough estimate of the coefficient of thermal conductivity, one can use the formula

$$\varkappa \sim Cl^{\text{eff}} v, \tag{1}$$

where C is the heat capacity per unit volume, v is the average velocity of the quasiparticles. In the very low temperature region $l^{\text{eff}} \sim d$ and κ increases with temperature. At higher temperatures, (but for $T \ll \Theta$) κ decreases exponentially. Therefore the temperature dependence of the coefficient of thermal conductivity is bell-shaped (see Fig. 1). If we disregard details, then these qualitative considerations are well-confirmed by experiment (see, for example, the article by Berman[2]).

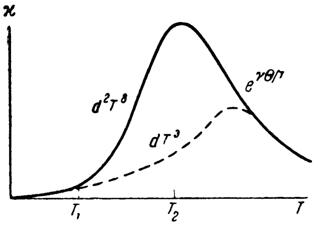

FIG. 1

However, the validity of these ideas from a theoretical point of view was placed in doubt by Pomeranchuk.[3] He noted that if only three-phonon processes are considered, then it follows from the appropriate conservation laws that long wavelength longitudinal phonons can only collide with phonons of comparatively long wavelengths. As a result the scattering probability for longitudinal phonons goes to zero so rapidly that the coefficient of thermal conductivity diverges. But this difficulty turns out to be connected to a considerable extent with the assumption made by Pomeranchuk[3] about the isotropy of the phonon spectrum. As Herring showed,[4] if anisotropy and degeneracy of the phonon spectra are considered, then for the majority of crystals this divergence is not present, or it leads to unimportant dimensional effects in real crystals. We note that in general similar questions do not arise for the case of spin waves having a quadratic dispersion law.

Another difficulty with the theory of the thermal conductivity of dielectrics is related to the fact that one is not able to solve the kinetic equation in this case (even for $T \ll \Theta$). In order to obtain qualitative results a relaxation time (see, for example, [3]) is usually introduced by one method or another. In this connection, the normal collisions between quasiparticles which are not accompanied by Umklapp processes (N-processes) are, as a rule, not taken into consideration (see, however, the article by Klemens[5]). Such an approach is possibly reasonable at not too low temperatures, when the probabilities for N-processes and Umklapps (U-processes) are of the same order ($l^N \sim l^U$), since the N-processes by themselves do not lead to thermal resistance. However, in the region of sufficiently low temperatures where $l^N \ll l^U$, N-processes obviously can be important. But it is just in this case that one can systematically solve the kinetic equation (see the book by Peierls[6]). As the calculations show, the consideration of N-processes does not lead to a qualitative change in the results associated with volume collisions (U-processes[7] and scattering by impurities[8]).

The situation is different in the case when the thermal resistance is due to the scattering of quasiparticles by the boundaries of the sample (or by any kind of macroscopic distortions). It is clear that the following inequality will hold in a sufficiently massive and pure sample at sufficiently low temperatures:

$$l^N \ll d \ll l^V, \qquad (2)$$

where l^V is the effective mean free path which characterizes the volume collisions with loss of momentum. Here a situation arises which is completely analogous to that investigated earlier by the author for the case of electrons in a metal.[9] Moving like a particle undergoing Brownian motion, the phonon (or spin wave) travels between two collisions with path limits of order $d^2/l^N \gg d$. Therefore, according to (1),

$$\varkappa \sim Cvd^2/l^N,$$

i.e., the order of magnitude of the thermal conductivity coefficient and its dependence on the parameters T, d, and so forth are significantly altered by the N-collisions.

For a quantitative investigation of this mechanism it is natural to use a hydrodynamical approach, as was also done in [9]. The fact is that the N-processes only lead to internal equilibrium inside each volume element (large in comparison with l^N) which may move as a whole with arbitrary velocity u. Therefore it is clear in advance that with fulfillment of inequality (2) the distribution function will have the form

$$f_0(\varepsilon - \mathbf{pu}) \equiv \{[\exp((\varepsilon - \mathbf{pu})/T) - 1]\}^{-1},$$

where $\epsilon(\mathbf{p})$ is the energy of the quasiparticle, **p** is the quasimomentum.

The velocity $\mathbf{u}(\mathbf{r})$ of the ordered motion as a function of coordinates must satisfy a hydrodynamic equation of the Navier-Stokes type. Such an equation is derived below and is then used to investigate the thermal conductivity of dielectrics (see Sec. 2) and ferrodielectrics (see Sec. 3).

1. The hydrodynamic equation can be obtained starting from the kinetic equation in analogy to the way this is done for ordinary gases (see, for example, [10]). In the case of interest to us, it is convenient to write the linear kinetic equation for the distribution function $f(\mathbf{p}, \mathbf{r})$ in the form

$$\mathbf{v}\frac{\partial f}{\partial \mathbf{r}} + (\mathbf{v}\nabla T)\frac{\partial f_0}{\partial T} = \hat{J}^N f + \hat{J}^V f,$$

where $\mathbf{v} = \partial\epsilon/\partial\mathbf{p}$, the first term on the right side describes N-processes, and the second describes collisions accompanied by losses of quasimomentum.

For an approximate solution of this equation it is natural to take advantage of the fact that the term $\hat{J}^N f$ is large in comparison with the terms $\mathbf{v}\cdot\partial f/\partial\mathbf{r}$ and $\hat{J}^V f$. In fact the solution is sought in the form of a series in the small parameters l^N/d and l^N/l^V. The method of successive approximations leads, as one can easily show, to the following system of equations:

$$\hat{J}^N f^{(0)} = 0, \qquad \mathbf{v}\partial f^{(0)}/\partial\mathbf{r} = \hat{J}^N f^{(1)},$$

$$\mathbf{v}\frac{\partial}{\partial\mathbf{r}} f^{(1)} - (\mathbf{v}\nabla T)\frac{\varepsilon}{T} f_0'(\varepsilon) - \hat{J}^V f^{(0)} = \hat{J}^N f^{(2)};$$

$$f = f^{(0)} + f^{(1)} + f^{(2)} + \dots$$

It follows from the first equation that $f^{(0)} = f_0(\epsilon - \mathbf{p}\cdot\mathbf{u})$. The second equation, which now takes the form $v_i p_k f_0'(\epsilon)\,\partial u_k/\partial x_i = \hat{J}^N f^{(1)}$ cannot be solved in general. One can only prove that $f^{(1)}$ contains the derivatives $\partial u_k/\partial x_i$ linearly.

Since we are interested in the region of very low temperatures, and in the equation under investigation it is obvious that only energies $\epsilon \lesssim T$ are important, we can assume that the dispersion law of the quasiparticles is a power law [$\epsilon(\mathbf{p}) = s(\mathbf{n})p$ for phonons and $\epsilon(\mathbf{p}) = \alpha(\mathbf{n})p^2$ for spin waves, **n** is a unit vector in the direction of **p**]. In this connection $f^{(1)} = \psi_{ik}(\epsilon/T, \mathbf{n})A\partial u_i/\partial x_k$, as is easy to prove. The functions ψ_{ik} of dimensionless variables satisfy equations which do not contain small parameters; in the isotropic case $\psi_{ik} = n_i n_k \psi(\epsilon/T)$. The temperature-dependent coefficient A can be easily found in any specific case.

We then multiply the third equation by **p** and integrate. Since the integral on the right side is identically equal to zero, the sought-for equation for $\mathbf{u}(\mathbf{r})$ appears as a result. In the isotropic case it is convenient to write this equation in the form

$$\gamma\nabla T = \nu\Delta\mathbf{u} - \mathbf{u}/\tau^V; \tag{3}$$

$$\gamma \approx \frac{1}{\beta T}\int d\mathbf{p}\; pv\varepsilon f_0'(\varepsilon), \qquad \nu = v_T l^N \approx \frac{1}{\beta}\int d\mathbf{p}\; pv\psi,$$
$$\frac{\mathbf{u}}{\tau^V} \approx \frac{1}{\beta}\int d\mathbf{p}\,\mathbf{p}\,\hat{J}^V(\mathbf{up})\, f_0'(\varepsilon), \qquad \beta = \int d\mathbf{p}\; p^2 f_0'(\varepsilon). \tag{4}$$

In analogy with an ordinary gas, the kinematic viscosity ν is written in the form of the product of the average velocity v_T of the quasiparticles ($v_T \approx (\partial\epsilon/\partial p)_{p=p_T}$, $\epsilon(p_T) = T$) times the quantity l^N, having the meaning of mean free path for N-processes. To this equation one must add the boundary condition: $\mathbf{u} = 0$ on the boundaries of the sample and on macroscopic distortions, which corresponds to diffusion scattering.

As is clear from the derivation, the hydrodynamic equation retains the same structure in the case of an anisotropic dispersion law. But, of course, all coefficients will now be tensors. Thus, for example, the term $\nu\Delta u_i$ is replaced by $\nu_{iklm}\partial^2 u_k/\partial x_l \partial x_m$; however, it is essential that all $\nu_{iklm} \sim \nu$. Therefore in what follows we shall start from Eq. (3), claiming in this connection only correct orders of magnitude of quantities and their correct dependence on the basic parameters.

The heat current density

$$\mathbf{Q} = \frac{1}{h^3}\int d\mathbf{p}\; \mathbf{v}\varepsilon f_0(\varepsilon - \mathbf{pu}) \cong \frac{\mathbf{u}}{3h^3}\int d\mathbf{p}\; pv\varepsilon f_0'(\varepsilon)$$

depends on the coordinates, and in order to determine the thermal conductivity coefficient κ it is necessary to average the vector $\mathbf{Q}$ over the transverse cross section of the sample: $\overline{\mathbf{Q}} = \kappa\nabla T$. As an example we give the result of solving Eq. (3) in two elementary cases: a plate and a circular cylinder.

Let us represent the coefficient of thermal conductivity in the form $\kappa = Cl^{\text{eff}} v_T$, where

$$C = -\frac{1}{h^3 T}\int d\mathbf{p}\; \varepsilon^2 f_0'(\varepsilon)$$

is the heat capacity. Then for the plate*

$$l^{\text{eff}} \approx l^V\,(1 - z^{-1}\,\text{th}\, z)$$

and for the cylinder

$$l^{\text{eff}} \approx l^V\,[1 - I_1(z)/zI_0(z)], \qquad l^V = \tau^V v_T,$$
$$z = d/2\sqrt{l^V l^N}.$$

Here I_1 and I_0 are Bessel functions of imaginary argument while d is the thickness of the plate or the diameter of the cylinder.

In limiting cases both formulas lead, correct to within an unimportant numerical factor, to one and the same result: For $z \gg 1$, $l^{\text{eff}} \approx l^V$ and for $z \ll 1$, $l^{\text{eff}} \approx l^V z^2 \approx d^2/l^N$. This is natural since $z^2 \sim d^2/l^N l^V$ and the quantity d^2/l^N, as proved, has the meaning of effective mean free path for collisions with the boundaries. It is clear that the small difference of the results for plate and cylinder is not accidental in the hydrodynamical region. In what follows, by d we shall understand some characteristic transverse dimension of the sample (or the distance between macroscopic distortions).

2. Now we go on to concrete results for the phonon thermal conductivity of dielectrics. At low temperatures the interaction of phonons with each other occurs principally by means of three-phonon processes. Starting from the appropriate collision integral (see, for example, Peierls[6]) and the relations (4), one can prove that in order of magnitude $l^N_{pp} \sim a(Ms^2/\Theta_D)(\Theta_D/T)^5$ where a is the lattice constant, M is the atomic mass, s is the velocity of sound, Θ_D is the Debye temperature, $\Theta_D \sim hs/a$.

For three-phonon U-processes, analogously to the way this was done in[7], one can easily show that

$$l^U_{pp} = s\tau^U_{pp} \sim a\,(Ms^2/\Theta_D)\,(T/\Theta_D)^{5/2}\exp(\gamma\Theta_D/T), \quad \gamma = 1.$$

The numerical value of the parameter γ depends on the behavior of the phonon spectrum inside the boundaries of the whole Brillouin zone.[1] It is curious that in other respects the results given, also including the pre-exponential factor $(T/\Theta_D)^{5/2}$, do not depend on the nature of the dispersion law.

In not very pure samples scattering by impurities may also turn out to be important. The corresponding mean free path $l_{pi} \sim (a/c)(\Theta_D/T)^4$, c is the concentration of impurity atoms; here $(l^V)^{-1} = (l^U_{pp})^{-1} + (l_{pi})^{-1}$.

An approximate curve of the temperature dependence of the thermal conductivity coefficient for a dielectric is shown in Fig. 1. In the region of very low temperatures, as long as $l^N_{pp} \gg d$ it is obvious that $l^{\text{eff}} \sim d$ and $\kappa \approx C_p ds \approx da^{-3}s\,(T/\Theta_D)^3$. Then, beginning with the temperature T_1 at which $l^N_{pp} \sim d$ up to the temperature T_2 at which $d^2/l^N \sim l^V$, the diffusion mechanism plays the major role and

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*th = tanh.

[1] $\gamma\Theta$ denotes the smallest possible value of the energy of the colliding phonons, $\epsilon(\mathbf{p}_1) + \epsilon(\mathbf{p}_2)$, allowable by the conservation laws: $\epsilon(\mathbf{p}_1) + \epsilon(\mathbf{p}_2) = \epsilon(\mathbf{p}_3)$, $\mathbf{p}_1 + \mathbf{p}_2 - \mathbf{p}_3 = \mathbf{b}$, $\mathbf{b}$ is a reciprocal lattice vector.

$$\varkappa \approx C_p\,(d^2/l_{pp}^N)\,s \approx (\Theta_D\,/\,Ms)(d^2/a^4)\,(T/\Theta_D)^8.$$

Then the usual decrease of the thermal conductivity begins according to either an exponential law (if U-processes dominate) or according to a 1/T law (if scattering by impurities plays the major role). The usual temperature dependence $\kappa(T)$ is also shown in Fig. 1 (the dotted curve) for comparison.

As follows from inequality (2) the diffusion mechanism can appear only in a sufficiently massive sample of dielectric. It is difficult to obtain any reliable estimate for the thickness d, but apparently it would be sufficient if d were ~ 1 cm.

3. Spin waves are basically responsible for heat transport in ferrites at low temperatures. Actually the heat capacity of a spin wave gas, $C_S \approx a^{-3}(T/\Theta_C)^{3/2} \gg C_p$ if $T \ll \Theta_D^2/\Theta_C$. In addition, N-collisions between spin waves are considerably more probable than N-collisions of spin waves with phonons (see the review article by Akhiezer et al.[11]). Therefore for the investigation of this temperature region, in which the diffusion mechanism dominates, one can disregard the presence of phonons.

Collisions between spin waves are connected with relativistic and exchange interactions. The mean free path for the corresponding N-processes in order of magnitude equals (see [11])

$$l_{ss}^{(r)} \sim a\,(\Theta_C/\mu M_0)^2\,[1 + \exp(\beta\mu M_0/T)], \quad l_{ss}^{(e)} \sim a\,(\Theta_C/T)^{7/2},$$

where μ is the Bohr magneton, M_0 is the saturation magnetic moment per unit volume, β is the anisotropy constant. The external magnetic field is assumed to be weak ($\mu H \ll T$).

From the expressions given, it follows that the diffusion mechanism must appear even in relatively thin samples of ferrite. In particular, it is sufficient if $d \gg a(\Theta_C/\mu M_0) \sim 10^{-3}$ to 10^{-4} cm. If this inequality is satisfied, then the coefficient of thermal conductivity behaves as follows (see Fig. 2). At very low temperatures, as long as $d \ll l_{ss}^{(r)}$, $l^{\mathrm{eff}} \approx d$ and $\kappa \approx C_S d v_T \approx (\Theta_C d/ha^2) \times (T/\Theta_C)^2$. Then in the interval from

$$T_1[l_{ss}^{(r)}(T_1) \sim d] \text{ to } T_2[d^2/l_{ss}^{(e)}(T_2) \sim l_s^V(T_2)]$$

the thermal conductivity is determined by the diffusion mechanism and

$$\varkappa \approx \frac{\Theta_C}{h}\frac{d^2}{a^3}\left(\frac{T}{\Theta_C}\right)^2$$

$$\times \begin{cases} (\mu M_0/\Theta_C)^2 \exp(-\beta\mu M_0/T), & T_1 \ll T \ll \mu M_0, \\ (\mu M_0/\Theta_C)^2, & \mu M_0 \ll T \ll \Theta_C(\mu M_0/\Theta_C)^{4/7}, \\ (T/\Theta_C)^{7/2}, & \Theta_C(\mu M_0/\Theta_C)^{4/7} \ll T \ll T_2. \end{cases}$$

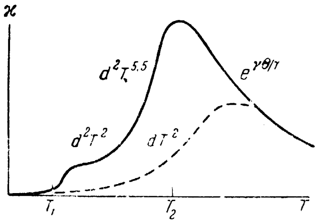

FIG. 2

For $T > T_2$ in a sufficiently pure sample κ decreases exponentially. The impurity thermal resistance essentially depends on the magnetic properties of the impurity atoms (see the article by Bar'yakhtar and Urushadze[12]).

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