INFLUENCE OF HIGH-ORDER ANHARMONICITY ON THE THERMAL CONDUCTIVITY OF FERRODIELECTRICS

R. N. GURZHI and I. N. OLEĬNIK

Physico-technical Institute, Ukrainian Academy of Sciences; Institute of Radioelectronics, Ukrainian Academy of Sciences

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It was previously shown by one of the authors that under certain conditions (perfect bulk samples and low temperatures) the thermal resistance of a ferrodielectric is determined by Umklapp processes associated with collisions involving the participation of an arbitrarily large number of magnons.[2] The present article is devoted to the further investigation of this mechanism. It is shown that in the case when $\Theta_D \ll \Theta_C$ (Θ_D is the Debye temperature, Θ_C is the Curie temperature), high-order magnon-magnon collisions are important only at sufficiently low temperatures whereas at higher temperatures collisions between phonons play the major role. Mixed processes in which two phonons and a large number of magnons participate may give a significant contribution in the region of intermediate temperatures. It is also shown that under the conditions considered a constant magnetic field has a very substantial effect on the thermal conductivity of a ferrodielectric, where this effect already appears in relatively weak fields $\mu H \sim T\sqrt{T/\Theta_C}$.

1. AS is well known, the transport of heat in dielectrics is accompanied by a directed flow of quasiparticles which are created at the "hot" end of the sample and disappear at the "cold" end. The thermal resistance of the dielectric is therefore determined by collisions in which the total quasimomentum of the quasiparticles is not preserved. Umklapp collisions (U-processes) play the principal role in sufficiently perfect and massive samples. It is well known that at low temperatures the probability of an U-process is exponentially small, and normal collisions in which the quasimomentum is conserved occur much more frequently. By virtue of the frequent normal collisions, the momentum which is lost by one of the quasiparticles as a result of an Umklapp process is redistributed among all the quasiparticles. Therefore the thermal resistance of the dielectric is determined by the total number of U-processes in the system, independently of whether these processes are possible for all of the quasiparticles (see Peierls[1]).

Usually in calculating the thermal conductivity one takes into account the U-processes associated with collisions involving the participation of the minimum possible number of quasiparticles. (For example, three for collisions between phonons and four or three for magnon-magnon collisions.) However, one of the authors of the present article has shown that such a method of treatment may turn out to be incorrect in that case when the energy of the quasiparticles, as a function of the magnitude of the quasimomentum, varies more rapidly than according to a linear law.[2]

For spin waves having a quadratic dispersion law, the calculation based on the Holstein–Primakoff representation leads to the following expression for the total number of U-processes:[2]

$$N^U = \sum_{n\geq 2} A_n (T/\Theta_c)^{3n-2} \exp\left(-\frac{\pi^2\Theta_c}{nT}\right). \qquad (1)$$

Here Θ_C is the Curie temperature; n is the number of magnons before and after the collision (only the exchange interaction, which conserves the number of magnons, was taken into account); the A_n are numerical coefficients whose dependence on n turns out to be of little importance.

From the cited formula it is clear that with a reduction of the temperature, collision processes of increasingly higher order will play the major role in the quantity N^U. We emphasize that this assertion is essentially based on only two assumptions: a) The "nonlinearity" of the dispersion law, b) the presence in the interaction Hamiltonian of the quasiparticles of anharmonicities of different order.

According to Eq. (1) the dependence of the coefficient of thermal conductivity of a ferrodielectric on the temperature, $\kappa(T) \sim 1/N^U(T)$, has the shape of a wavy curve shown in Fig. 1 (the lower bell-shaped curve). The decrease of $\kappa(T)$ according to the law $T^{11/2}$ for $T < T_0$ is due to the influence of the boundaries in the presence of the frequent normal collisions.[3] (If the normal collisions are not taken into consideration, then in this region $\kappa \sim T^2$.) We note that very similar results have recently been obtained experimentally by Tsarev[4,5] in connection with investigations of the thermal conductivity of the ferrodielectric $CrBr_3$.

The present article is devoted to an investigation of the effect, which is more detailed than what was done earlier,[2] with certain circumstances which arise under actual experimental conditions taken into account. The influence of phonons (Sec. 2) and of a constant magnetic field (Sec. 3) on the thermal conductivity of ferrodielectrics will be treated below.

2. For collisions between phonons, high-order processes obviously cannot be important. (Although it cannot be excluded that, with a reduction of the temperature, the four-phonon process becomes more probable

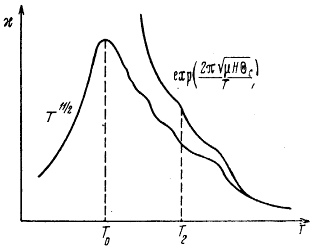

FIG. 1

than the three-phonon process.) Therefore it is clear beforehand that at sufficiently low temperatures the number N^U of U-processes will be determined by magnon-magnon collisions of high order. However, if the Debye temperature $\Theta_D \ll \Theta_C$, then phonon-phonon U-processes will dominate at higher temperatures (but temperatures such that $\exp(\Theta_D/T) \gg 1$). In addition, the question arises as to the role of mixed processes, in which several phonons and a large number of magnons participate.

As usual, let us write down the Hamiltonian of the system in terms of boson operators a and c for the phonons and magnons. We have

$$\mathscr{H} = \mathscr{H}_0 + \sum \Phi_{3p} a_\lambda^+ a_\mu^+ a_\nu + \sum_{n\geq 2} \Phi_{2ns} c_1^+ \ldots c_n^+ c_{n+1} \ldots c_{2n}$$
$$+ \sum_{n\geq 1} \Phi_{2ns,1p}\, a_\lambda^+ c_1^+ \ldots c_n^+ c_{n+1} \ldots c_{2n} + \sum_{n\geq 1} \Phi_{2ns,2p}\, a_\lambda^+ a_\mu c_1^+ \ldots c_n^+ c_{n+1} \ldots c_{2n}. \quad (2)$$

Here $\hat{\mathscr{H}}_0$ is the gas Hamiltonian, the second term describes the usual interaction between phonons, and the third term describes many-magnon processes related to the exchange interaction; finally the last two terms describe mixed processes involving the participation of one or two phonons. (These terms can be obtained from the exchange Hamiltonian by means of an expansion with respect to small displacements of the atoms in the lattice.[6]) As the analysis showed, mixed processes involving the participation of three or more phonons give a small contribution. The dependence of the amplitude Φ on the momenta of the magnons has an extremely cumbersome form (see, for example, the expression for Φ_{6s} given in [2]) and is not essential for what follows.

Starting from the Hamiltonian (2), it is not difficult to derive the system of kinetic equations for the phonon distribution function $N^p = \langle a^+ a \rangle$ and for the magnon distribution function $N^s = \langle c^+ c \rangle$. In this connection, just as in [2], we confine our attention to the Born approximation, in which the kernels of the collision integrals are proportional to $|\Phi|^2$ for the appropriate process. (One can show that the higher-order terms in perturbation theory give small contributions.)

The kinetic equations have the form

$$(\mathbf{v}^\sigma \nabla T) \frac{\partial N_0(\varepsilon^\sigma)}{\partial T} = \sum_{\sigma'} (\hat{I}_N^{\sigma\sigma'} N^{\sigma'} + \hat{I}_U^{\sigma\sigma'} N^{\sigma'}). \quad (3)$$

Here $\mathbf{v}^\sigma = \partial\epsilon^\sigma/\partial\mathbf{p}$, $N_0(\epsilon) = [\exp(\epsilon/T) - 1]^{-1}$, $\epsilon(\mathbf{p})$ is the energy of the quasiparticle, $\mathbf{p}$ is its quasimomentum, $\hat{I}_N$ is the operator describing the normal collisions, and $\hat{I}_U$ is the operator for Umklapp collisions; the summation goes over all branches of the energy spectrum of the phonons and of the magnons (σ = p, s).

The solution of the system of Eqs. (3) can be obtained without difficulty if, following Peierls,[1] we utilize the fact that at low temperatures normal collisions occur much more frequently than Umklapp collisions. In this connection the distribution function both for the phonons and for the magnons has the form $N_0(\epsilon - \mathbf{p}\cdot\mathbf{u}) \approx N_0(\epsilon) - \mathbf{p}\cdot\mathbf{u}\, N_0'(\epsilon)$, and the drift velocity $\mathbf{u}$ is determined by the relation

$$-\frac{1}{3}\nabla T \sum_\sigma \int d\mathbf{p}(\mathbf{p}\mathbf{v}^\sigma) \frac{\partial N_0(\varepsilon^\sigma)}{\partial T} = u_i \sum_{\sigma,\sigma'} \int d\mathbf{p}\cdot\mathbf{p}\hat{I}_U^{\sigma\sigma'} p_i N_0'(\varepsilon^{\sigma}). \quad (4)$$

In the linear approximation the heat flux $\mathbf{Q}$ is proportional to the drift velocity:

$$\mathbf{Q} = -\mathbf{u} \sum_\sigma \frac{1}{3h^3} \int d\mathbf{p}(\mathbf{p}\mathbf{v}^\sigma)\varepsilon^\sigma N_0'(\varepsilon^\sigma). \quad (5)$$

According to Eqs. (4) and (5) we have the following result for the thermal conductivity tensor ($Q_i = -\kappa_{ik}\partial T/\partial x_k$),

$$\varkappa_{ik} = (\hat{\alpha}^{-1})_{ik} \frac{T}{h^3} \Big[\sum_\sigma \frac{1}{3} \int d\mathbf{p}(\mathbf{p}\mathbf{v}^\sigma)\varepsilon^\sigma N_0'(\varepsilon^\sigma)\Big]^2,$$
$$\alpha_{ik} = \sum_{\sigma,\sigma'} \int d\mathbf{p} p_i \hat{I}_U^{\sigma\sigma'} p_k N_0'(\varepsilon^{\sigma'}). \quad (6)$$

It is obvious that the tensor α_{ik} is proportional to the total number of U-processes in the system. We also note that, in order of magnitude the expression standing inside the square brackets in formula (6) is equal to the sum of the phonon and magnon thermal conductivities. (For arbitrary dispersion laws $\mathbf{p}\cdot\mathbf{v} \approx \epsilon(\mathbf{p})$.) As a result, to within numerical coefficients of slight importance one can represent the coefficient of thermal conductivity in the form

$$\varkappa \sim \frac{T^2 a^5 (C_s + C_p)^2}{\hbar\Theta_C N^U} \quad (7)$$

Here C_s and C_p are the magnon and phonon thermal conductivities, respectively, a is the lattice constant, and the number of U-processes is given by

$$N^U = \sum_{n,m,\mathbf{b}\neq 0} \int B(\mathbf{p}_1, \ldots, \mathbf{p}_{n+m}) \exp\Big\{-\frac{1}{2T}\Sigma_1^{n+m}\Big\} \delta(\Sigma_1^n - \Sigma_{n+1}^{n+m})$$
$$\times \delta\Big(\sum_1^n \mathbf{p}_i - \sum_{n+1}^{n+m} \mathbf{p}_i + \mathbf{b}\Big) d\mathbf{p}_1 \ldots d\mathbf{p}_{n+m}, \quad (8)$$
$$\Sigma_m^n = \sum_m^n \varepsilon_i.$$

The quantities B are proportional to the squares of the matrix elements for the corresponding processes and they have an extremely complicated form; the $\mathbf{b}$ are the reciprocal lattice vectors.

Each term in N^U gives the number of U-processes owing to collisions in which n particles turn into m particles, where these particles can be of different types. At low temperatures the integrand in N^U is exponentially small and reaches its greatest value at a certain point $\{\mathbf{p}_{i0}\}$, at which Σ_1^{n+m} has the smallest value compatible with the laws for the conservation of energy and quasimomentum. Expanding the difference $\sum_1^{n+m} - (\sum_1^{n+m})_{min}$ in the neighborhood of this point and taking the conservation laws into consideration, we obtain a homogeneous form of $3(n+m)-4$ independent variables in the exponential function. The coefficients B can be taken outside of the integrals at the point $\{p_{io}\}$. As a result each term in N^U turns out to be proportional to the expression

$$T^{3/2(n+m)-2}\exp\left[-\frac{1}{2T}(\Sigma_1^{n+m})_{min}\right].$$

We note that the power of the temperature in the pre-exponential factor does not depend on the dispersion laws of the quasiparticles and is determined only by the order of the process, n + m, whereas the argument of the exponential function is essentially related to the form of the dispersion laws.

Omitting the simple but extremely cumbersome calculations, which are analogous to those carried out by Peierls,[1] we cite the result for the simplest case of isotropic power-law dispersion laws:

$$\varepsilon^p(\mathbf{p}) = \Theta_D\frac{a}{\hbar}p, \quad \varepsilon^s(\mathbf{p}) = \Theta_C\left(\frac{a}{\hbar}p\right)^2.$$

Correct to within numerical coefficients of slight significance, we have[1)]

$$N^U \approx \alpha x^{5/2}e^{-1/2x} + \sum_{n\geqslant 2}(n!)^2x^{3n-2}e^{-\beta/nx} + \alpha\left(\sum_{n=1}^{[\beta]}(n!)^2x^{3n-1/2}\right.$$
$$\times\exp\left\{-\frac{1}{x}\frac{n}{8\beta}\left(1+\frac{2\beta}{n}\right)^2\right\} + \sum_{n=[\beta]}^{[4\beta]}(n!)^2x^{3n-1/2} \tag{9}$$
$$\left.\times\exp\left\{-\frac{2}{x}\left(1-\frac{1}{4}\sqrt{\frac{n}{\beta}}\right)^2\right\}\right) + \alpha^2\sum_{n=1}^{[4\beta]}(n!)^2x^{3n+1}\exp\left\{-\frac{1}{2x}\left(1-\frac{n}{8\beta}\right)\right\}$$

where $\alpha = \Theta_D/Ms^2$, $x = T/2\pi\Theta_D$, $\beta = \pi\Theta_C/2\Theta_D$, and $[\ldots]$ denotes the integer part. The first two terms in this formula describe pure processes, and the remaining two describe mixed processes involving the participation of one or two phonons (compare with the Hamiltonian (2)).

The analysis of expression (9) shows that in the region of relatively high temperatures (but $\exp(1/x) \gg 1$) the mixed terms are small in comparison with the first (purely phonon) term, but at low temperatures the second term (purely magnon) is always the major term. Mixed processes can give an appreciable contribution to N^U only in the region of intermediate temperatures, where generally speaking all of the terms in (9) are of the same order.

Let us neglect the mixed processes and we shall assume the dispersion law of the phonons to be arbitrary.[2)] Then

$$N^U \approx \alpha x^{5/2}e^{-\gamma/2x} + \sum_{n\geqslant 2}(n!)^2x^{3n-2}e^{-\beta/nx}, \tag{10}$$

where $\gamma \sim 1$. (In the case of a linear dispersion law for the phonons $\gamma = 1$, but for a sinusoidal law $\gamma = 2/\pi$.) We note that at sufficiently low temperatures, a small group of terms with numbers[7]

$$n_{extr} \approx \pi\left[\frac{2T}{\Theta_C}\ln\left(\frac{\Theta_C}{T}\right)\right]^{-1/2}$$

will play the major role in the summation over n.

From formulas (9) and (10) it is clear that the nature of the temperature dependence of the coefficient of thermal conductivity is primarily determined by the relationship between the parameters $\gamma \approx 1$ and β.

For $\Theta_C \gg \Theta_D$ ($\beta \gg \gamma$) the condition for the dominance of phonon U-processes over magnon processes has the form $n_{extr}(T) < 4\beta/\gamma$, or

$$T > T_1 \approx \Theta_D^2\gamma^2/16\Theta_C\ln(\Theta_C/\Theta_D).$$

It is also not difficult to verify that $C_s(T_1) \sim C_p(T_1)$. Therefore, for $T \gg T_1$ the thermal conductivity is entirely determined by the phonons and

$$\varkappa_p \approx \frac{T}{\hbar a}\frac{Ms^2}{\Theta_D}\left(\frac{T}{\Theta_D}\right)^{9/2}\exp\left(\frac{\gamma\pi\Theta_D}{T}\right), \quad T \gg T_1, \tag{11}$$

and for $T \ll T_1$ the magnon U-processes with numbers $n > [4\beta/\gamma] \gg 1$ are dominant and (see [7])

$$\varkappa \sim \exp\left(2\pi\sqrt{2\frac{\Theta_C}{T}\ln\frac{\Theta_C}{T}}\right).$$

In the opposite limiting case, $\Theta_C \ll \Theta_D$, the phonon contribution is unimportant and the results of [2] are valid:

$$\varkappa_s \approx \frac{T}{\hbar a}\left(\frac{T}{\Theta_C}\right)^6\left[\sum_{n\geqslant 2}(n!)^2\left(\frac{T}{\Theta_C}\right)^{3n}\exp\left(-\frac{\pi^2\Theta_C}{nT}\right)\right]^{-1}. \tag{12}$$

3. A constant magnetic field leads to the appearance of an activation energy in the magnon spectrum: $\epsilon^s(p) = \Theta_C(ap/\hbar)^2 + \mu H$. As a result in expression (9) for the number of U-processes each term, describing a collision involving the participation of magnons, obtains a correction in the exponential which is proportional to the order n of the process: $-\mu Hn/T$. (The pre-exponential factors remain unchanged because for processes with conservation of the number of magnons the quantity μH drops out of the law of energy conservation.) From here it follows that the presence of a magnetic field can only lead to a decrease of the contribution N^U of the mixed processes in comparison with purely phonon processes, and it does not change the relation between mixed and purely magnon processes. Therefore, just as above we shall not take the mixed processes into account.

The magnetic field turns out to have an influence on the coefficient of thermal conductivity for two reasons. In the first place, it leads to a decrease of the probability of magnon-magnon U-processes. We note that at sufficiently low temperatures this effect will appear even in very weak fields, when $\mu H n_{extr} \sim T$, that is, for $\mu H \sim T(T/\Theta_C)^{1/2}$. In the second place, the magnetic field affects the spin thermal conductivity C_s appearing in formula (7), where C_s becomes exponentially small for $\mu H \gg T$.

Omitting the simple calculations, we present the expressions for the coefficient of thermal conductivity $\kappa(T, H)$ in different limiting cases.

[1)]For the sake of brevity, pre-exponential factors of the type $(\Theta_D/\Theta_C)^{3n}$, π^{3n}, etc. have been omitted in the expression given here; these factors do not turn out to have a significant influence on the nature of the temperature dependence of the coefficient of thermal conductivity.

[2)]The assumption about the quadratic nature of the magnon spectrum in the present case is justified in [2]. For simplicity we shall also regard it as isotropic, having a cubic crystal in mind.

For $\Theta_D \gg \Theta_C$ and $H \ll H_1 = \Theta_D^2\gamma^2/4\mu\Theta_C$ (the opposite inequality corresponds to unrealistically large fields) we have

$$\varkappa \sim \begin{cases} \varkappa_s \exp\left(\frac{\pi\mu H}{T}\sqrt{\frac{\Theta_c}{2T}\left(\ln\frac{\Theta_c}{T}\right)^{-1}}\right) = \\ = \varkappa_s e^{H/H_3}, \quad T \gg T_2 \text{ or } H \ll H_2; \\ C_p{}^2 \exp\left(\frac{2\pi}{T}\sqrt{\mu H \Theta_c}\right) = \\ = C_p{}^2 \exp\left(\frac{\sqrt{HH_2}}{H_3}\right), \quad T \ll T_2 \text{ or } H \gg H_2. \end{cases}$$

In the case $\Theta_D \ll \Theta_C$ the temperature dependence of κ for a fixed magnetic field $H \ll H_1$ has the form

$$\varkappa \sim \begin{cases} \varkappa_p, & T \gg T_1, \\ \varkappa_s \exp\left(\frac{\pi\mu H}{T}\sqrt{\frac{\Theta_c}{2T}\left(\ln\frac{\Theta_c}{T}\right)^{-1}}\right), & T_2 \ll T \ll T_1, \\ C_p{}^2 \exp\left(\frac{2\pi}{T}\sqrt{\mu H \Theta_c}\right), & T \ll T_2, \end{cases}$$

and the dependence of κ on the magnetic field for a fixed temperature $T \ll T_1$ will be given by

$$\varkappa \sim \begin{cases} \varkappa_s \exp(H/H_3), & H \ll H_2, \\ C_p{}^2 \exp(\sqrt{HH_2}/H_3), & H_2 \ll H \ll H_1, \\ \varkappa_p, & H \gg H_1. \end{cases}$$

For $H \gg H_1$ and $T \gg T_1$ we have $\kappa = \kappa_p$.

In these formulas $\kappa_s(T)$ and $\kappa_p(T)$ denote the magnon and phonon thermal conductivities for $H = 0$ (see formulas (11) and (12)) and the following notation is adopted:

$$T_1 = \frac{\Theta_D{}^2\gamma^2}{16\Theta_C \ln(\Theta_C/\Theta_D)}, \quad T_2 = \frac{\mu H}{4\ln(\Theta_C/\mu H)}, \quad \mu H_1 = \frac{\Theta_D{}^2\gamma^2}{4\Theta_C},$$

$$\mu H_2 = 8T \ln\frac{\Theta_C}{T}, \quad \mu H_3 = \frac{\sqrt{2}}{\pi} T\left(\frac{T}{\Theta_C}\ln\frac{\Theta_C}{T}\right)^{1/2}.$$

The dependence of the coefficient of thermal conductivity on the temperature and on the magnetic field for the case $\Theta_D \gg \Theta_C$ is shown in Figs. 1 and 2. The lower bell-shaped curve in Fig. 1 corresponds to $H = 0$.

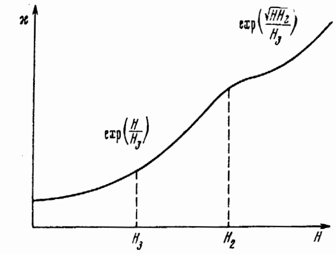

FIG. 2

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