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EFFECT OF ANHARMONICITIES OF HIGH ORDER ON TRANSPORT PROCESSES IN SOLIDS

R. N. GURZHI

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It is shown that at sufficiently low temperatures transport processes in solids may be determined by anharmonicities of arbitrarily high order. For this it is necessary that the energy of the corresponding quasiparticles vary more rapidly than linearly as a function of the quasimomentum. A specific calculation is carried out for the spin-wave induced thermal conductivity of ferrites.

As is well-known, finite coefficients of thermal and electrical conductivities can be obtained for an infinite, perfect single-crystal only by taking Umklapp processes into account. If these processes are absent, collisions between quasiparticles do not change their total quasimomentum, which will increase indefinitely with time under the action of external forces. If the quasiparticles obey Bose statistics (for example, phonons or spin waves), then at low temperatures the probability of a collision accompanied by an Umklapp process is exponentially small [$\sim \exp(-\gamma\Theta/T)$, where γ is a numerical coefficient and Θ is the Debye temperature in the case of phonons or the Curie temperature in the case of spin waves; $T \ll \Theta$]. In order to understand the origin of this exponential dependence, we consider as an example the case when there are two particles, both before and after the collision.

From the laws of energy and momentum conservation

$$\varepsilon_1 + \varepsilon_2 - \varepsilon_3 - \varepsilon_4 = 0, \qquad \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3 - \mathbf{k}_4 = \mathbf{b}$$

(b = reciprocal lattice vector) we can see that at least two of the four k vectors must be of order b. The energy corresponding to these large k is $\epsilon \sim \Theta \gg T$. Therefore the probability of a collision will be proportional to $\exp\{-(\epsilon_1 + \epsilon_2)/T\}$, where it is necessary to take the minimum value allowable by the conservation laws for the energy $\epsilon_1 + \epsilon_2$ of the colliding particles; this minimum is evidently $\sim\Theta$. The probability of a similar process in which not necessarily two but some arbitrary number of particles, n, participate is proportional to $\exp(-T^{-1}\Sigma\epsilon_i)$, where one must take for $\Sigma\epsilon_i$ the smallest value allowed by the conservation laws:

$$\sum_1^n \varepsilon_i - \sum_{n+1}^{2n} \varepsilon_i = 0, \qquad \sum_1^n \mathbf{k}_i - \sum_{n+1}^{2n} \mathbf{k}_i = \mathbf{b}.$$

In addition, the power of the small parameter (T/Θ) in the factor preceding the exponential will increase with an increase of the order of the process according to the law $(T/\Theta)^{\beta n}$; the constant β depends on the nature of the dispersion law. It is easy, however, to understand that the exponential factors, which primarily determine the small probability of collisions accompanied by Umklapp processes, may increase with increase of the order of the process n. For this to happen, it is necessary that the energy of a quasiparticle change more rapidly, as a function of the quasimomentum, than according to a linear law.

As a specific example, we consider spin waves in a cubic crystal. Then, at not too low temperatures[1] we have $\epsilon(k) = \Theta(ak)^2$, the minimum nonzero value is $|b| = 2\pi/a$ and, as is easily shown, $\beta = 3$; here Θ is of the order of magnitude of the Curie temperature and a is the lattice constant. If one assumes that the quadratic dispersion law is valid up to $k \sim b/2n$, then the minimum value of $\Sigma\epsilon_i$ is reached at the point $k_i = b/2n$ $(i \le n)$ or $k_i = -b/2n$ $(i > n)$ and equals $n\epsilon(b/2n) = \pi^2\Theta/n$. Therefore one would expect that the total probability for loss of momentum by the spin wave system, with account of anharmonicities of all orders, would be proportional to an expression of the form

$$\sum_n B_n (T/\Theta)^{3n} \exp(-\pi^2\Theta/nT),$$

where the B_n are numerical coefficients.

It follows from this formula that with a lowering of the temperature the kinetic processes will be determined by anharmonicities of ever increasing order. We note that the latter assertion is essentially based on only two assumptions: the "nonlinear" dispersion law and the presence of anharmonicities of different order in the quasiparticle interaction Hamiltonian.

In order to show how similar results can be specifically obtained by starting from a given Hamiltonian, we calculate the thermal conductivity of a ferrite, due to spin waves. In the simplest case of a cubic crystal, the Hamiltonian for the exchange interaction has the form

$$\mathcal{H} = \frac{\alpha}{2}\sum_{k} k^2 \mathbf{m}_{\mathbf{k}}\mathbf{m}_{-\mathbf{k}}, \qquad (1)$$

where $\alpha = a^2\,\Theta/\mu M_0$, μ is the Bohr magneton, M_0 is the saturation magnetic moment, and $\mathbf{m}_{\mathbf{k}}$ is the Fourier component of the magnetization $\mathbf{m}(\mathbf{r})$ corresponding to wave vector $\mathbf{k}$. In the following we shall assume that the external magnetic field is negligibly small ($\mu H \ll T$) and that relativistic interactions are unimportant ($T \gg \mu^2/a^3 \sim 1°\mathrm{K}$).

As usual, following Holstein and Primakoff, we express the magnetization $\mathbf{m}(\mathbf{r})$ in terms of boson operators $a^+(\mathbf{r})$ and $a(\mathbf{r})$:

$$m_+(\mathbf{r}) = m_x + im_y$$
$$= (2\mu M_0)^{1/2}\, a^+(\mathbf{r})\,[1 - \mu a^+(\mathbf{r})\, a(\mathbf{r})/2M_0]^{1/2},$$
$$m_-(\mathbf{r}) = m_x - im_y = (m_+(\mathbf{r}))^+, \quad m_z = M_0 - \mu a^+(\mathbf{r})\, a(\mathbf{r}).$$

We expand the square root appearing in these formulas in a series with respect to $a^+(\mathbf{r})\,a(\mathbf{r})$, and we go over to the Fourier representation. As a result, the Hamiltonian for the exchange interaction (1) can be represented in the form

$$\hat{\mathcal{H}} = \sum \varepsilon(k)\, a^+_{\mathbf{k}} a_{\mathbf{k}} + \sum \Phi_{12,34}\, a^+_1 a^+_2 a_3 a_4$$
$$+ \sum \Phi_{123,456}\, a^+_1 a^+_2 a^+_3 a_4 a_5 a_6 + \ldots, \qquad (2)$$

$\epsilon(k) = \Theta(ak)^2$; the real and symmetric amplitudes Φ can be easily found, for example

$$\Phi_{123,456} = \frac{1}{32}\Theta a^2 \left(\frac{\mu}{M_0 V}\right)^2 \Big[\sum_{i=1}^{6} \mathbf{k}_i^2$$
$$- \frac{1}{9}\Big(\sum_{i=1}^{6}\mathbf{k}_i\Big)^2\Big]\, \Delta(\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3 - \mathbf{k}_4 - \mathbf{k}_5 - \mathbf{k}_6),$$

where V is the volume of the sample.

The kinetic equation for the spin wave distribution function, $N_{\mathbf{k}} = \langle a^+_{\mathbf{k}} a_{\mathbf{k}}\rangle$, can be easily obtained. Omitting the standard calculations, we present only the result. The collision operator $\hat{W}$ in the quadratic approximation (with respect to the amplitudes Φ) can be represented in the form of a sum of operators associated with individual terms in the Hamiltonian (2):

$$\hat{W} = \sum \hat{W}_n.$$

If the nonequilibrium correction to the distribution function is represented in the form

$$N_{\mathbf{k}} - N_k^{(0)} = N_k^{(0)}(1 + N_k^{(0)})\,\varphi_{\mathbf{k}}, \quad N_k^{(0)} = [1 - \exp(\varepsilon_k/T)]^{-1},$$

then in the linear (in $\Phi_{\mathbf{k}}$) approximation

$$\hat{W}_{2n} N_{\mathbf{k}_1} = -n\,(n!)^2 \frac{2\pi}{\hbar}\sum |\Phi_{1,\ldots,n;\,n+1,\ldots,2n}|^2\, \delta\Big(\sum_1^n \varepsilon_i - \sum_{n+1}^{2n}\varepsilon_i\Big)$$
$$\times \exp\Big(-\frac{1}{T}\sum_1^n \varepsilon_i\Big)\prod_1^{2n}(1 + N_i^{(0)})$$
$$\times \Big(\sum_1^n \varphi_i - \sum_{n+1}^{2n}\varphi_i\Big)\,\Delta\Big(\sum_1^n \mathbf{k}_i - \sum_{n+1}^{2n}\mathbf{k}_i - \mathbf{b}\Big),$$

where the summation is over all wave vectors $\mathbf{k}_2, \mathbf{k}_3, \ldots \mathbf{k}_{2n}$ (within the Brillouin zone) and over all reciprocal lattice vectors $\mathbf{b}$.

For what follows, it is convenient to represent the collision operator as the sum of two operators, $\hat{W}^N$ and $\hat{W}^U$, which describe processes in which quasimomentum is conserved ($\mathbf{b} = 0$) and Umklapp processes ($\mathbf{b} \neq 0$). In this connection, the kinetic equation is written in the form

$$N_k^{(0)}(1 + N_k^{(0)})\,\varepsilon_k T^{-2}(\mathbf{v}_{\mathbf{k}}\nabla T) = \hat{W}^N\varphi_{\mathbf{k}} + \hat{W}^U\varphi_{\mathbf{k}}. \qquad (3)$$

The left-hand side determines the change in the number of spin waves due to the effect of a small temperature gradient ∇T; $\mathbf{v}_{\mathbf{k}} = \hbar^{-1}\,\partial\epsilon_{\mathbf{k}}/\partial\mathbf{k}$.

At low temperatures the term $\hat{W}^U\varphi_{\mathbf{k}}$ is small. It is natural to take advantage of this fact in order to approximately solve the kinetic equation.[2,3] However, if we simply discard the term $\hat{W}^U\varphi_{\mathbf{k}}$ in the zero-order approximation, then the resulting equation does not have a solution. We therefore look for a solution of the kinetic equation in the form $\varphi_{\mathbf{k}} = \varphi_{\mathbf{k}}^{(0)} + \varphi_{\mathbf{k}}^{(1)}$, where $|\varphi_{\mathbf{k}}^{(0)}| \gg |\varphi_{\mathbf{k}}^{(1)}|$ and the following equations are satisfied

$$\hat{W}^N\varphi_{\mathbf{k}}^{(0)} = 0, \quad \hat{W}^N\varphi_{\mathbf{k}}^{(1)} = N_k^{(0)}(1 + N_k^{(0)})\,\varepsilon_k T^{-2}(\mathbf{v}_{\mathbf{k}}\nabla T) - \hat{W}^U\varphi_{\mathbf{k}}^{(0)}.$$

From the first equation $\varphi_{\mathbf{k}}^{(0)} = \mathbf{g}\mathbf{k}$, and the constant vector $\mathbf{g}$ can be found from the condition for solvability of the second equation with respect to $\varphi_{\mathbf{k}}^{(1)}$:

$$\sum_{\mathbf{k}} \mathbf{k} N_k^{(0)}(1 + N_k^{(0)})\,\frac{\varepsilon_k}{T^2}(\mathbf{v}_{\mathbf{k}}\nabla T) = \sum_{\mathbf{k}}\mathbf{k}\hat{W}^U\mathbf{g}\mathbf{k}.$$

For a simple cubic lattice, taking into account only the minimum absolute values of $\mathbf{b}$ ($|\mathbf{b}| = 2\pi/a$ and, as is easily shown, quantities of higher order in $\exp(-\Theta/T)$ can be neglected), we obtain

$$\mathbf{g} = \mathbf{R}/A, \quad \mathbf{R} = \frac{1}{(2\pi)^3}\int \mathbf{k} N_k^{(0)}(1 + N_k^{(0)})\,\frac{\varepsilon_k}{T^2}(\mathbf{v}_{\mathbf{k}}\nabla T)\, d\mathbf{k},$$

$$A = \left(\frac{2\pi}{a}\right)^3 \frac{\pi}{\hbar}\sum_{n=2}^{\infty}(n!)^2 \frac{1}{(2\pi)^{6n}}\int d\mathbf{k}_1 \ldots d\mathbf{k}_{2n}\,|\Phi_{1,2,\ldots,n;n+1,\ldots,2n}|^2$$
$$\times\,\delta\Big(\sum_1^n \varepsilon_i - \sum_{n+1}^{2n}\varepsilon_i\Big)\exp\Big(-\frac{1}{T}\sum_1^n\varepsilon_i\Big)\prod_1^{2n}(1 + N_i^{(0)})$$
$$\times\,\delta\Big(\sum_1^n \mathbf{k}_i - \sum_{n+1}^{2n}\mathbf{k}_i - \frac{2\pi}{a}\boldsymbol{\nu}\Big), \qquad (4)$$

where ν is an arbitrary unit vector.

The integrals in the last formula can be approximately evaluated since, for $T \ll \Theta$, a small region in k-space surrounding the point at which $\Sigma_1^n \epsilon_i$ reaches the minimum value allowed by the conservation laws gives, evidently, the major contribution. In order to find these minimum values, it is necessary to know the dispersion law for $\epsilon(\mathbf{k})$ for large values of $|\mathbf{k}|$. It may be possible, for example, to use the law

$$\varepsilon(\mathbf{k}) = 2\Theta(3 - \cos k_x a - \cos k_y a - \cos k_z a),$$

but if we simply assume $\epsilon(k) = \Theta(ak)^2$, then the corresponding minimum values are reached at $|k_i| = \pi/na$ (see the beginning of the article). Hence the assumed quadratic nature of the dispersion law becomes all the more valid as the number n increases, but even for the evaluation of the first integral (n = 2) this assumption, as one can easily verify, does not lead to a large error.

Omitting the rather cumbersome calculations, we present at once the result for the coefficient of thermal conductivity κ (the heat current $Q = \Sigma N_{\mathbf{k}} v_{\mathbf{k}} \epsilon(\mathbf{k}) = \kappa \nabla T$):

$$\varkappa \cong \frac{T}{\hbar a}\left[\sum_{n\geqslant 2} B_n \left(\frac{T}{\Theta}\right)^{3n-6} \exp\left(-\frac{\pi^2}{n}\frac{\Theta}{T}\right)\right]^{-1},$$

$$B_n \cong \pi^{11-3n} 2^{5-6n} \frac{(n!)^2}{n^3} s^{2n-2}, \qquad (5)$$

where $s = a^3 M_0/\mu$ is the atomic spin; the summation goes from $n = 2$ up to $n \sim \Theta/T$.

The last limitation is essential, for owing to the presence of the factors $(n!)^2$ in the coefficients B_n the terms of the series begin to increase for $n > \Theta/T$. This is not surprising since it has been assumed in the calculation of the integrals in (4) that $\Theta/nT \gg 1$. The exact value of the upper limit on the summation over n is not important, since for $n \sim \Theta/T$ the terms of the series are negligibly small. Moreover, it turns out that at any temperature only one specific term of the sum in (5), the term with

$$n_T \sim \pi\left(\frac{\Theta}{3T\ln(\Theta/T)}\right)^{1/3} \ll \frac{\Theta}{T}$$

plays a major role (it is assumed that $\ln(\Theta/T) \gg 1$).

As a result, the following picture is obtained for the temperature dependence of the coefficient of thermal conductivity. At relatively high temperatures (but for $e^{\Theta/T} \gg 1$) the first term of the sum plays the dominant role, in agreement with [3],

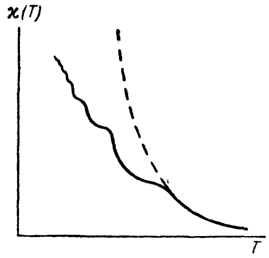
$$\varkappa \cong (T/\hbar a)\, e^{\pi^2\Theta/2T}. \qquad (6)$$

Beginning at the temperature $T_1 \sim \Theta/10$, the second term (n = 3) of the sum dominates, and the temperature dependence becomes more gradual:

$$\varkappa \sim (T/\hbar a)\,(\Theta/T)^3 e^{\pi^2\Theta/3T}.$$

Finally, for $\ln(\Theta/T) \gg 1$, when the term with number $n_T \gg 1$ plays the major role,

$$\varkappa \sim \frac{T}{\hbar a}\left(\frac{\Theta}{T}\right)^6 \exp\left(2\pi\sqrt{3}\sqrt{\frac{\Theta}{T}\ln\frac{\Theta}{T}}\right).$$



The temperature dependence of $\kappa(T)$ is qualitatively shown in the figure (solid curve). The upper dashed curve corresponds to the usual dependence (6).

In order to clarify the possibility of observing such a dependence, we shall determine the effective free path length in the region where deviations from (6) begin, i.e., at $T \sim T_1$. Comparing the well-known formula

$$\varkappa \sim Clv$$

(C is the heat capacity, l is the effective mean free path, and v is the average velocity; in the present case $v \sim \hbar^{-1} \partial\epsilon/\partial k|_{\epsilon \sim T} \sim a\sqrt{T\Theta}/\hbar$) with the results obtained above, one can show that the function $l(T_1) \sim 1$ cm for $s \sim 1$. Therefore, it is necessary to have pure single-crystal samples[1] of transverse dimension d > 1 cm. Otherwise, it appeared that boundary scattering of spin waves would play the major role.

However, in reality the transfer of momentum from the spin-wave gas to the walls may be strongly impeded, owing to normal collisions in which momentum is conserved. We denote the free path lengths corresponding to normal collisions and Umklapp collisions by l^N and l^U, respectively. Estimates show that the inequality $l^N \ll d \ll l^U$ will be fulfilled in not too thin samples at a sufficiently low temperature. In this connection a situation arises, analogous to that considered earlier by the author for the case of an electron gas, [4] and the effective free path turns out to be of order d^2/l^N and is, therefore, considerably larger than d. These considerations, which may also turn out to be important for the

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[1] We note that at low temperatures the scattering of spin waves by impurity atoms is unimportant in sufficiently pure samples, since the long-wavelength oscillations are weakly scattered by local defects.

phonon thermal conductivity, will be examined separately.

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