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Kirill D. Belashchenko

Kurchatov Institute, belashchenko@unl.edu

D V Livanov

Moscow Institute of Steel and Alloys

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Phonon effects in the thermoelectric power of impure metals

K D Belashchenko[†] and D V Livanov[‡]

[†] Russian Research Center, ‘Kurchatov Institute’, Moscow 123182, Russia

[‡] Moscow Institute of Steel and Alloys, Leninskii Prospect 4, Moscow 117936, Russia

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Abstract. Using the quantum transport equations for interacting electrons and phonons we study the phonon effects in the thermoelectric transport in impure metals. The contributions of both equilibrium phonons (the diffusive part) and non-equilibrium phonons (the drag effect) are investigated. We show that the drag effect which dominates in the thermopower of pure samples is strongly suppressed even by a small impurity concentration owing to the inelastic electron–impurity scattering processes. As a result we find the form of the phonon drag thermopower of an impure metal taking into account both normal and Umklapp processes of the electron–phonon scattering.

1. Introduction

The thermoelectric transport properties of metals are difficult to calculate and measure compared with the electrical transport properties. From the theoretical point of view this difficulty is because of the compensative nature of the thermoelectricity. Indeed, the thermoelectric effect in metals is a small result of two opposing currents owing to electrons and holes which almost completely cancel. The surviving small difference strongly depends upon the electron band structure, the phonon spectrum and the peculiarities of the scattering mechanisms. The measurable quantity is the thermopower, or the Seebeck coefficient, $S = -\beta/\sigma$, where σ and β are the conductivity and the thermoelectric coefficient, respectively (defined as $\mathbf{j} = \sigma \mathbf{E} + \beta \nabla T$, where \mathbf{j} , \mathbf{E} and ∇T are the electric current, external electric field and temperature gradient, respectively). In the most simple scenario the thermopower of a normal metal contains two contributions. The first of them is the diffusive part S_d connected with the expansion of the energy-dependent electronic parameters near the Fermi level by the parameter T/E_F (T is the temperature, and E_F is the Fermi energy). The diffusive part of the thermopower is negative for electrons and positive for holes. The second relevant term is the so-called phonon drag contribution which is induced only by the non-equilibrium correction to the phonon distribution function and has the order $(T/\Theta_D)^3$ (Θ_D is the Debye temperature, and we consider the low-temperature case $T \ll \Theta_D$). It is clear that the phonon drag contribution S_g can be ignored only at rather low temperatures $T \ll (\Theta_D/E_F)^{1/2} \Theta_D$. S_g has the same sign as S_d for normal electron–phonon processes, and the opposite sign for Umklapp electron–phonon processes. Thus, the total sign of the phonon drag contribution is conditioned by the form of the Fermi surface, which governs whether the normal or the Umklapp processes dominate. The interplay of the two terms S_d and S_g can explain the features of the thermopower of pure metals [1].

The situation with dirty metallic systems and alloys is much more complicated because the mechanism of the influence exerted by impurities on the phonon drag was not well understood until now. It is clear from the qualitative point of view that S_g is maximal when electrons are the main scatterers for phonons, while the activation of other phonon scattering mechanisms results in the rapid suppression of the drag effect. At high temperatures $T \simeq \Theta_D$ the phonon–phonon scattering plays the essential role leading to the $1/T$ decrease in β_g with the temperature growth [1, 2]. At low temperatures the effective rate of the phonon–phonon scattering falls off as q_T^3 ($q_T \sim T/u$ is the characteristic wavevector of a thermal phonon, and u is the sound velocity). Moreover, the scattering of long-wave phonons on each other conserves the total wavevector, and hence such processes cannot be responsible for the relaxation of the total quasi-momentum of the phonon subsystem. This relaxation requires the Umklapp processes, and their probability falls off exponentially at low temperatures as $\exp(-\Delta_0/T)$, where Δ_0 has the order of the Debye temperature Θ_D . In dirty metals or alloys the direct phonon scattering on impurities with its probability proportional to $c_{imp}q_T^4$ (c_{imp} is the impurity concentration) can also become sufficient for the suppression of the phonon drag contribution. At the same time, the scattering rate owing to this mechanism quickly decreases with the decrease of temperature and cannot be responsible for the low-temperature suppression of S_g in dirty systems. We have shown that this suppression may take place at lower impurity concentrations than those required for the effective direct phonon–impurity scattering [3]. This fact is the result of the processes of inelastic scattering of electrons on impurities with the emission or absorption of a phonon accompanied by the dissipation of the total momentum of the phonon subsystem. This inelastic electron–impurity scattering also makes an important contribution to the conductivity of an impure metal at low temperatures together with other electron–phonon–impurity interference processes [4, 5].

To estimate the relative change of the phonon drag contribution to the thermopower of an impure system one must take into account that at low temperatures the ratio of the phonon emission or absorption rate owing to the inelastic electron–impurity scattering to the rate of ‘pure’ phonon–electron scattering is of the order $(q_T l)^{-1}$, where l is the electron mean free path [5]. Thus, the relative correction to the phonon drag contribution to the thermopower S_g (the ‘drag thermopower’) owing to such processes assumes the following form

$$\frac{S_g(c_{imp}) - S_g(0)}{S_g(0)} \sim -\frac{1}{q_T l} \sim -c_{imp} \frac{\Theta_D}{T}. \quad (1)$$

Many aspects of the phonon effects in the thermoelectric transport have been widely discussed. Phonons can affect the thermoelectric power in metals in two ways. The first is the influence of virtual phonons with the characteristic wavevector of the order of the Debye one. These phonons renormalize the thermoelectric power similarly to the electron mass enhancement [6, 7]. The second way is connected with thermal (real) phonons which affect the diffusive component (inducing the corrections similar to Bloch contribution to conductivity) and are also responsible for the drag component. The electron–phonon contribution to the diffusive thermopower was studied quasi-classically in a number of works (see, e.g., [8, 9]). The drag contribution to the thermopower was studied theoretically using the Boltzmann equation in the relaxation time approximation in [10, 11] (for a 3D metal) and in [12, 13] (for a 2D metal) and experimentally in [14–16] and very recently in [17]. In an impure metal at sufficiently low temperatures the electron–phonon scattering interferes with the impurity scattering generating the additional corrections both to the diffusive and to the drag thermopower. In this regime the quasi-classical approach becomes inadequate and one should use the system of quantum kinetic equations for electrons and phonons.

The role of the thermal phonons in the thermopower of impure 3D metallic systems at low temperatures is discussed in the present paper.

The paper is organized as follows. In section 2 we formulate the quantum transport equations for the interacting electron–phonon system with phonons supposed to be non-equilibrium. In section 3 we study the phonon-induced corrections to the diffusive component of the thermopower. The phonon drag component in impure metals is studied in section 4. Finally, in section 5 we compare different contributions and discuss the experimental situation.

2. Kinetic equations for electrons and phonons

In this section we describe the formalism that can be used to investigate the electron–phonon kinetics of normal metals. This formalism is based on the diagrammatic technique for non-equilibrium systems developed by Keldysh [18] where the Green functions of electrons and phonons, as well as the electron self-energy and the polarization operator are represented by matrices:

$$\begin{aligned}\widehat{G} &= \begin{pmatrix} 0 & G^A \\ G^R & G^C \end{pmatrix} & \widehat{D} &= \begin{pmatrix} 0 & D^A \\ D^R & D^C \end{pmatrix} \\ \widehat{\Sigma} &= \begin{pmatrix} \Sigma^C & \Sigma^R \\ \Sigma^A & 0 \end{pmatrix} & \widehat{\Pi} &= \begin{pmatrix} \Pi^C & \Pi^R \\ \Pi^A & 0 \end{pmatrix}.\end{aligned}\quad (2)$$

Here the indices A and R stand for advanced and retarded functions, respectively, while the diagonal components G^C and D^C contain the information about the statistical distribution of electrons and phonons. The kinetic equation for electrons was derived in [4, 18] where thermal equilibrium was assumed for the phonon subsystem. However, it is well known that the thermoelectric response is governed by two equally important contributions, the diffusive thermopower and the drag effect. The former is connected with the electron–hole asymmetry, and the latter is the contribution of the non-equilibrium of the phonon subsystem. Indeed, if the phonon–electron interaction is not small compared with other mechanisms of phonon scattering, the phonons are ‘dragged’ by the non-equilibrium electrons producing the phonon thermal current as a response to the external electric field. Hence in order to describe the phonon drag contribution to the thermopower one has to write a separate kinetic equation for phonons and solve it together with the kinetic equation for electrons. The quasi-classical limit of this system of kinetic equations was studied in a number of works (see, e.g., [19]). At high temperatures the quasi-classical approach is quite sufficient for the description of the electron–phonon kinetics. However, at low temperatures the quantum many-particle (interaction interference) effects become significant and must be taken into account. For example, as we will see below, at sufficiently low temperatures the processes of the phonon emission or absorption connected with the inelastic scattering of electrons on impurities dominate over the phonon–phonon and direct phonon–impurity scattering. Hence one should use a quantum analogue of the well known kinetic equation for phonons to describe the dependence of the drag effect on temperature and impurity concentration at low temperatures.

The derivation of the quantum kinetic equation for phonons follows that of the kinetic equation for electrons [4, 18]. We take the non-perturbed phonon Green function in the following form

$$D_\lambda^R(\omega, \mathbf{q}) = [D_\lambda^A(\omega, \mathbf{q})]^* = \frac{\omega_\lambda(\mathbf{q})}{2} \left(\frac{1}{\omega - \omega_\lambda(\mathbf{q}) + i0} - \frac{1}{\omega + \omega_\lambda(\mathbf{q}) + i0} \right) \quad (3)$$

where $\omega_\lambda(\mathbf{q})$ is the dispersion law for the given branch of the phonon spectrum denoted as λ . First we must write down the Dyson's equation for the phonon Green function in two ways:

$$D_0^{-1}(x_2)\widehat{D}(x_2, x_1) = \widehat{\sigma}_x \omega_\lambda^2(-i\nabla_2) \left[\delta(x_2 - x_1) + \int d^4y \widehat{\Pi}(x_2, y)\widehat{D}(y, x_1) \right] \quad (4a)$$

$$D_0^{-1}(x_1)\widehat{D}(x_2, x_1) = \omega_\lambda^2(i\nabla_1) \left[\delta(x_2 - x_1) + \int d^4y \widehat{D}(x_2, y)\widehat{\Pi}(y, x_1) \right] \widehat{\sigma}_x \quad (4b)$$

where $x = (t, \mathbf{r})$, $D_0^{-1}(x) = -\partial^2/\partial t^2 - \omega_\lambda^2(-i\nabla)$, and $\widehat{\sigma}_x$ is the known Pauli matrix. The equations (2) must be written separately for each branch of the phonon spectrum; the corresponding indices of the phonon Green function and polarization operator are omitted. For acoustic phonons in the Debye approximation $\omega_\lambda^2(-i\nabla) = -u_\lambda^2 \Delta$, where u_λ is the sound velocity and Δ is the Laplacian operator. Since phonons do not carry electric charge, the Dyson's equation for phonons does not contain the potentials of the electromagnetic field.

Taking the difference of the 22-components of (4a) and (4b), entering new variables $X = (x_1 + x_2)/2 = (t, \mathbf{R})$ and $x = x_2 - x_1 = (\theta, \mathbf{r})$ and performing the Fourier transformation over the relative coordinates

$$\widehat{D}(\omega, \mathbf{q}, X) = \int d\theta d\mathbf{r} \widehat{D}(\theta, \mathbf{r}, X) \exp(i\theta\omega - i\mathbf{r}\mathbf{q}) \quad (5)$$

we obtain the kinetic equation for phonons

$$\begin{aligned} -2i \left[\omega \frac{\partial}{\partial t} + \frac{\partial \omega_\lambda^2(\mathbf{q})}{\partial \mathbf{q}} \nabla_{\mathbf{R}} \right] D^C(\omega, \mathbf{q}, \mathbf{R}, t) &= \omega_\lambda^2(\mathbf{q}) [\Pi^C(D^R - D^A) - D^C(\Pi^R - \Pi^A)] \\ &+ \frac{i}{2} \omega_\lambda^2(\mathbf{q}) \{D^R + D^A, \Pi^C\} + \frac{i}{2} \omega_\lambda^2(\mathbf{q}) \{D^C, \Pi^R + \Pi^A\} \\ &+ \frac{i}{2} \{ \omega_\lambda^2(\mathbf{q}), \Pi^C D^A + \Pi^R D^C \} + \frac{i}{2} \{ \omega_\lambda^2(-\mathbf{q}), D^R \Pi^C + D^C \Pi^A \}. \end{aligned} \quad (6)$$

Here we introduced the Poisson brackets arising from the non-homogeneity of the system (see [4])

$$\{f, g\} = \left(\frac{\partial f}{\partial \omega} \frac{\partial g}{\partial t} - \frac{\partial g}{\partial \omega} \frac{\partial f}{\partial t} \right) + \left(\frac{\partial f}{\partial \mathbf{R}} \frac{\partial g}{\partial \mathbf{q}} - \frac{\partial g}{\partial \mathbf{R}} \frac{\partial f}{\partial \mathbf{q}} \right). \quad (7)$$

In equilibrium $D^C = h_0(\omega)(D^R - D^A)$ where $h_0(\omega) = \coth(\omega/2T)$. In the general case this component can be presented in the following form

$$D^C = h(\omega, \mathbf{q}, X)(D^R - D^A) + \frac{i}{2} \{D^R + D^A, h\}. \quad (8)$$

The left-hand side of (6) may be expressed as $i\{D^C, \omega^2 - \omega_\lambda^2(\mathbf{q})\}$. Substituting (8) into (6) we obtain

$$\begin{aligned} i\{h, \omega^2 - \omega_\lambda^2(\mathbf{q})\}(D^R - D^A) &= \omega_\lambda^2(\mathbf{q}) [\Pi^C - h(\Pi^R - \Pi^A) + \frac{i}{2} \{h, \Pi^R + \Pi^A\}](D^R - D^A) \\ &+ \frac{i}{2} \omega_\lambda^2(\mathbf{q}) \{D^R + D^A, \Pi^C - h(\Pi^R - \Pi^A)\} \\ &+ \frac{i}{2} \{ \omega_\lambda^2(\mathbf{q}), \Pi^C - h(\Pi^R - \Pi^A) \} (D^R - D^A). \end{aligned} \quad (9)$$

The last two terms in (9) are quadratic with respect to the external perturbation and may be neglected in the theory of linear response. Finally, we obtain

$$i\{h, \omega^2 - \omega_\lambda^2(\mathbf{q})\} = \omega_\lambda^2(\mathbf{q}) [\Pi^C - h(\Pi^R - \Pi^A) + \frac{i}{2} \{h, \Pi^R + \Pi^A\}]. \quad (10)$$

This is the general kinetic equation for phonons. The coordinate derivatives in the Poisson brackets reflect the influence of the temperature gradient: $\partial/\partial \mathbf{R} = \nabla T \partial/\partial T$. If there is no temperature gradient, all Poisson brackets vanish.

The kinetic equation for phonons is very similar to the known kinetic equation for electrons derived in [4]

$$i\{\epsilon - \epsilon(\mathbf{p}), S\} = \Sigma^C - S(\Sigma^A - \Sigma^R) + \frac{1}{2}i\{\Sigma^A + \Sigma^R, S\}. \quad (11)$$

Here ϵ is the electron energy, $\epsilon(\mathbf{p})$ is the electron dispersion function (we assume the single-band spectrum, but the generalization of (11) for the multi-band case is straightforward), and $S(\epsilon, \mathbf{p}, X)$ is the electron distribution function (in equilibrium $S_0(\epsilon) = -\tanh(\epsilon/2T)$). Equations (10) and (11) form a closed system for the variable functions h and S . The electron distribution function S enters (10) through the polarization operator $\widehat{\Pi}$, while the phonon function h enters (11) through the electron self-energy $\widehat{\Sigma}$.

The thermoelectric coefficients may be calculated in two equivalent ways. The first way is to find the thermal current as a response to the external electric field, and the second way is to calculate the electric current generated by the applied temperature gradient. The first approach is more simple for the calculation of the drag effect because of the vanishing of all Poisson brackets in the phonon kinetic equation

$$\Pi^C - h(\omega, \mathbf{q})(\Pi^R - \Pi^A) = 0. \quad (12)$$

To find the drag contribution to the thermopower one must substitute the phonon distribution function h obtained from (12) into the expression for the phonon thermal current

$$\mathbf{Q}_{ph} = \sum_{\mathbf{q}} \frac{d\omega(\mathbf{q})}{d\mathbf{q}} \omega(\mathbf{q}) b_{\mathbf{q}}^+ b_{\mathbf{q}} = \frac{i}{2} \int \omega \frac{d\omega_{\mathbf{q}}}{d\mathbf{q}} \frac{D^C(\omega, \mathbf{q}, \mathbf{E})}{\omega_{\mathbf{q}}} \frac{d^3q}{(2\pi)^4} \quad (13)$$

where $b_{\mathbf{q}}^+$ and $b_{\mathbf{q}}$ are the phonon creation and annihilation operators; the thermal current (13) is connected with the drag thermoelectric coefficient β_g as $\mathbf{Q}_{ph} = -\beta_g T \mathbf{E}$.

The second approach is more convenient for the calculation of the phonon corrections to the diffusive thermopower. Here we present the main formulae for this procedure.

The electron kinetic equation for a system with the temperature gradient has the following form

$$\mathbf{v} \nabla T \frac{\partial S}{\partial T} = I(S) \quad (14)$$

where the collision integral

$$I(S) = -i[\Sigma^C - S(\Sigma^A - \Sigma^R)] + \frac{1}{2}i\{\Sigma^A + \Sigma^R, S\}. \quad (15)$$

Note that the components of the self-energy in the first term of equation (15) also contain the corrections in the form of the Poisson brackets.

The electron distribution function is found by iterations

$$S(\epsilon, \mathbf{p}) = S_0(\epsilon) + \phi_0(\epsilon, \mathbf{p}) + \phi_1(\epsilon, \mathbf{p}) \quad (16)$$

where ϕ_0 is the term generated by the elastic impurity scattering and ϕ_1 is the contribution of the phonon scattering processes (or any other interactions), together with the phonon renormalization of the impurity scattering integral. The elastic impurity scattering in our model is described by a short-range isotropic electron-impurity potential $V_{e-imp} = V_0 \delta(\mathbf{r})$. In this approximation $\phi_0(\epsilon, \mathbf{p}) = -\tau \mathbf{v} \nabla T \partial S_0/\partial T$, where

$$\tau = (\pi v(0) N_{imp} |V_0|^2)^{-1} \quad (17)$$

is the momentum relaxation time, $\nu(0) = mp_F/\pi^2$ is the single-electron density of states at the Fermi level (m is the electron mass and p_F is the Fermi momentum), and N_{imp} is the number of impurity atoms per unit volume. The value V_0 has the order of $\nu(0)^{-1}$. It is assumed that $|\phi_0| \gg |\phi_1|$, i.e. the impurity scattering is much stronger than all other scattering mechanisms (see also the discussion in section 4).

The electric current

$$\mathbf{j} = 2e \int \mathbf{v} S(\epsilon, \mathbf{p}) \text{Im} G^A(\epsilon, \mathbf{p}) \frac{d^3 p d\epsilon}{(2\pi)^4} \quad (18)$$

is connected with the diffusive thermoelectric coefficients β_d and β_g as $\mathbf{j} = \beta_d \nabla T_{el} + \beta_g \nabla T_{ph}$, where ∇T_{el} and ∇T_{ph} are the temperature gradients in the electron and phonon subsystems, respectively. Solving the kinetic equation for electrons assuming the phonon subsystem to be in equilibrium, we can obtain the diffusive contribution to the thermoelectric coefficient β_d . Note that the factor G^A in (18) also contains the phonon-induced contribution: $\delta_{ph} G^A(S) = G_0^{A2} \Sigma_{ph}^A(S)$ where Σ_{ph}^A is the contribution of the phonon processes into the electron self-energy.

Below we will use the kinetic equations to describe the influence of the phonons and impurities on the diffusive thermopower (section 3) and on the phonon drag effect (section 4).

3. Phonon corrections to the diffusive thermopower of an impure metal

Similarly to the phonon effects in the conductivity of an impure metal [4, 5] one can expect to observe two corrections to the diffusive thermoelectric coefficient: the one connected with the pure electron–phonon interaction, and the interference correction induced by the inelastic scattering of electrons on impurities with the emission or absorption of a phonon.

Below we will calculate the phonon corrections to the diffusive thermopower following the procedure described in section 2. To this end, we will find the electric current as a response to the applied temperature gradient assuming that the phonon subsystem is maintained in equilibrium. The application of the quantum kinetic equation formalism to the thermopower problem was described in detail in [7]. However, we emphasize that here we are interested in the role of thermal phonons with the characteristic wavevector q_T , but not the phonon renormalization of the thermoelectric coefficient by virtual phonons studied in [7]. Therefore, in the calculation of the thermoelectric coefficient we will be interested in the terms containing the imaginary part of the phonon propagator $\text{Im} D^R(\omega, \mathbf{q})$.

The diagrams for the electron self-energy describing the contribution of both the pure electron–phonon interaction (the analogue of the Bloch–Grüneisen correction to the conductivity of an impure metal) and the interference processes (including the inelastic scattering of electrons on impurities) coincide with those relevant to the calculation of the corrections to the conductivity [5]. Some of these diagrams are shown in figure 1. The vertices of pure electron–phonon scattering g_{ij}^k and of inelastic electron scattering on impurities γ_{ij}^k have the following form (see [5]; note the difference in the definition of the phonon propagator):

$$g_{ij}^k = i g_\lambda(\mathbf{q}) K_{ij}^k \quad g_\lambda(\mathbf{q}) = \frac{2E_F}{3\sqrt{MN}} \frac{\mathbf{q} \mathbf{e}_\lambda}{\omega_\lambda(\mathbf{q})} \quad \gamma_{ij}^k = i \frac{V_0}{\sqrt{MN}} \frac{(\mathbf{p} - \mathbf{p}') \mathbf{e}_\lambda}{\omega_\lambda(\mathbf{q})} K_{ij}^k \quad (19)$$

where $K_{ij}^1 = 2^{-1/2} \delta_{ij}$, $K_{ij}^2 = 2^{-1/2} (\sigma_x)_{ij}$, M is the ion mass, N is the number of elementary cells per unit volume, \mathbf{p} and \mathbf{p}' are the initial and final electron momenta, respectively, and \mathbf{e}_λ is the phonon polarization vector.

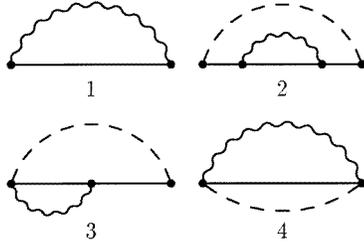


Figure 1. Some diagrams for the electron self-energy. Full curves are the electron propagators, wiggly curves are the phonon propagators, and broken curves represent the impurity scattering.

Diagram 1 in figure 1 describes the pure electron–phonon interaction, while other diagrams relate to the interference processes. As we will see below, the contribution of the interference diagrams to the thermopower is usually much less than that of the drag effect. Therefore, some diagrams of this type were omitted in figure 1 (for the full set of diagrams, see [5]). Diagram 1 gives the usual electron–phonon collision integral $I_0(\mathbf{p}, \epsilon)$ if one ignores all Poisson brackets in equation (15)

$$I_0(\epsilon, \mathbf{p}) = -8 \int \frac{d^3q d\omega}{(2\pi)^4} |g_l(\mathbf{q})|^2 \text{Im} G_0^A(\epsilon + \omega, \mathbf{p} + \mathbf{q}) \text{Im} D_l^R(\omega, \mathbf{q}) R_l(\epsilon, \mathbf{p}, \omega, \mathbf{q}) \quad (20)$$

where

$$R_\lambda(\epsilon, \mathbf{p}, \omega, \mathbf{q}) = \frac{1}{4} \{h_\lambda(\omega, \mathbf{q}) [S(\epsilon, \mathbf{p}) - S(\epsilon + \omega, \mathbf{p} + \mathbf{q})] + S(\epsilon, \mathbf{p}) S(\epsilon + \omega, \mathbf{p} + \mathbf{q}) - 1\} \quad (21)$$

and the indices l in (20) relate to the longitudinal branch of the phonon spectrum. Assuming the phonons to be in equilibrium we determine the phonon correction to the electronic distribution function $\phi_1(\epsilon, \mathbf{p})$. Next we calculate the correction to the thermoelectric coefficient according to the general expression (18). Finally after integration over \mathbf{p} we obtain

$$\beta_{e-ph} = -\frac{2e\pi^2}{3} \int \frac{d^3q d\epsilon d\omega}{(2\pi)^5} |g_l(\mathbf{q})|^2 \text{Im} D_l^R(\omega, \mathbf{q}) \frac{\tau^2 v v_F}{q} S_0(\epsilon) \frac{\partial}{\partial T} R_0(\epsilon, \omega) \quad (22)$$

where v_F is the Fermi velocity, $R_0(\epsilon, \omega)$ is obtained from (21) with the substitution of the equilibrium functions S_0 and h_0 , and the derivative over T acts only on the electronic distribution functions. The integration of the last expression over ϵ and ω requires the usual expansion of the energy-dependent electron spectrum parameters near the Fermi level because the integrand is uneven in the zero approximation. The integration yields

$$\beta_{e-ph} = -\frac{4}{3} \zeta(5) e \alpha \frac{\tau^2 v_F T^4}{E_F u_l^2} \quad (23)$$

where $\zeta(x)$ is the Riemann zeta-function ($\zeta(5) \approx 1.03$) and $\alpha = |g_l(\mathbf{q})|^2 v(0)/2$ is the dimensionless constant of electron–phonon interaction. This is the contribution of the pure electron–phonon interaction to the diffusive thermopower; in this sense it is analogous to the Bloch–Grüneisen contribution to the conductivity of an impure metal [4, 7].

Next we will study the correction to the diffusive thermoelectric coefficient induced by the processes described by the interference diagrams as well as by the terms with Poisson brackets in the contribution from diagram 1. Note that the interference contribution is connected both with the longitudinal and the transverse phonons; the contribution of the latter

is partially described by diagram 4. It will be clear from the following that the interference contributions contain the same dependence on temperature and impurity concentration as the phonon drag contribution described in the next section, but are much less in magnitude by the parameter m/M . In order to estimate the magnitude of this correction we calculate the contribution to diagram 1 induced by the Poisson brackets in equation (15). Here the corrections to the self-energy are non-zero only due to the modification of G^C . We have

$$\delta\Sigma^A = \frac{1}{4} \int \frac{d^3q d\omega}{(2\pi)^4} \left(\mathbf{v} + \frac{\mathbf{q}}{m}, \nabla T \right) |g_l(\mathbf{q})|^2 \frac{\epsilon + \omega}{T} \frac{\partial S_0(\epsilon + \omega)}{\partial \epsilon} D_l^A(\omega, \mathbf{q}) \times [G^{A^2}(\epsilon + \omega, \mathbf{p} + \mathbf{q}) + G^{R^2}(\epsilon + \omega, \mathbf{p} + \mathbf{q})]. \quad (24)$$

Substituting the self-energy (24) into (18) and integrating over the electron momentum we obtain the interference correction to the thermoelectric coefficient

$$\beta_{e-ph-imp} = -\pi e \int \frac{d^3q d\epsilon d\omega}{(2\pi)^5} \text{Im} D_l^R(\omega, \mathbf{q}) |g_l(\mathbf{q})|^2 \frac{v\tau}{q^2} \frac{\epsilon + \omega}{T} S_0(\epsilon) \frac{\partial S_0(\epsilon + \omega)}{\partial \epsilon}. \quad (25)$$

Finally, expanding the integrand near the Fermi level and performing the remaining integrations we obtain

$$\beta_{e-ph-imp} = -\frac{2}{9} \zeta(4) e \alpha \frac{\tau T^3}{E_F u_l}. \quad (26)$$

Diagrams 2–4 in figure 1, as well as other diagrams of the same type, produce the contribution of the same order. Apart from the longitudinal phonons contribution, two diagrams including diagram 4 also contain the transverse phonons contribution which has the additional factor $(u_l/u_t)^3$ compared with (26).

4. Phonon drag effect

Here we will use the kinetic equations for electrons and phonons described in section 2 to find the dependence of the phonon drag contribution to the thermopower (the ‘drag’ thermopower) on temperature and impurity concentration.

It is quite clear that the drag thermopower is maximal if the phonons are scattered mainly by electrons, and the growth of the frequency of other phonon scattering processes (e.g., scattering on impurities and on other phonons) leads to the suppression of the drag effect. Indeed, for a system in the external electric field these processes, unlike the scattering on non-equilibrium electrons, tend to eliminate the angle-dependent part of the phonon distribution function responsible for the phonon thermal current.

As was discussed in section 1, the processes of phonon–phonon and direct phonon–impurity scattering cannot be responsible for the low-temperature suppression of the drag effect, but the processes of the inelastic scattering of electrons on impurities with the emission or absorption of a phonon are well suited for this role. Here we will describe the features of this suppression in detail.

We will calculate the phonon thermal current as a response to the external electric field assuming that electrons are scattered mainly by impurities (the so-called ‘weak Mattissen rule’ conditions). This regime is realized in the temperature range where the effective frequency of the electron scattering on phonons $\nu_{e-ph} \sim T^3/\Theta_D^2$ (see [19]) is small compared with that of the electron scattering on impurities $\nu_{e-imp} \sim \tau^{-1}$: $T\tau \ll (\Theta_D/T)^2$. The fact that the phonon scattering of electrons is negligible sufficiently simplifies the system of the kinetic equations for electrons and phonons, because the electron distribution function in this case does not depend on the phonon one.

As was noted above, at low temperatures we must take account of the pure electron–phonon interaction and inelastic scattering of electrons on impurities. For the considered system with constant temperature the phonon kinetic equation assumes the form of equation (12). According to [5], the vertex γ_{ij}^k of the inelastic electron scattering on impurities contains an additional factor of the order $1/ql$ compared with the vertex of pure electron–phonon scattering [5]. This means that these processes are important for phonons with $ql \lesssim 1$.

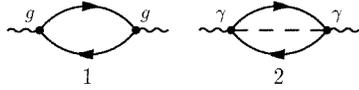


Figure 2. Diagrams for the polarization operator which contributes to the drag effect.

At first we will rewrite the kinetic equation neglecting the inelastic scattering of electrons on impurities (diagram 1 in figure 2):

$$|g_\lambda(\mathbf{q})|^2 \int \frac{d^3 p d\epsilon}{(2\pi)^4} R_\lambda(\epsilon, \mathbf{p}, \omega, \mathbf{q}) \operatorname{Im} G^A(\epsilon, \mathbf{p}) \operatorname{Im} G^A(\epsilon + \omega, \mathbf{p} + \mathbf{q}) = 0. \quad (27)$$

Let us introduce the standard representation of the distribution functions of electrons and phonons:

$$S(\epsilon, \mathbf{p}) = S_0(\epsilon) - \frac{\partial S_0(\epsilon)}{\partial \epsilon} \phi(\epsilon, \mathbf{p}) \quad h_\lambda(\omega, \mathbf{q}) = h_0(\omega) - \frac{\partial h_0(\omega)}{\partial \omega} \chi_\lambda(\omega, \mathbf{q}). \quad (28)$$

As long as we assume that the main process responsible for the electron momentum relaxation is the impurity scattering, the electron distribution function has the well known form

$$\phi(\epsilon, \mathbf{p}) \equiv \phi(\mathbf{p}) = e\tau \mathbf{v} \mathbf{E} \quad (29)$$

where e is the electron charge, $\mathbf{v} = d\epsilon_p/d\mathbf{p}$ is the electron velocity, and \mathbf{E} is the external electric field. After linearization the factor R in (27) assumes the following form

$$R_\lambda(\epsilon, \mathbf{p}, \omega, \mathbf{q}) = \frac{1}{4} [S_0(\epsilon + \omega) - S_0(\epsilon)] \frac{dh_0(\omega)}{d\omega} [\phi(\mathbf{p} + \mathbf{q}) - \phi(\mathbf{p}) - \chi_\lambda(\mathbf{q})]. \quad (30)$$

Assuming the single-band parabolic electron spectrum, in the absence of the Umklapp processes (27) is satisfied if $\chi(\mathbf{q}) = e\tau \mathbf{q} \mathbf{E}/m$, where m is the effective electron mass. Substituting this function into the formula for the phonon thermal current (13) we obtain the drag contribution to the thermoelectric coefficient β corresponding to the neglect of both the Umklapp processes and the inelastic electron scattering on impurities

$$\beta_g^0 = -\frac{2\pi^2}{45} \frac{e\tau T^3}{mu_l^3}. \quad (31)$$

This is the known result that may be obtained via solving the system of classical Boltzmann equations for electrons and phonons (see [19], §82).

Now let us consider the diagrams with the inelastic impurity scattering vertices. It is easy to see that in the combination of the distribution functions $R(\epsilon, \mathbf{p}, \omega, \mathbf{q})$ for diagram 2 the wavevector \mathbf{q} is replaced by the momentum \mathbf{p}' transferred to the impurity with the addition of the integration over \mathbf{p}' . From the other hand, the contribution of diagrams 3 and 4 is similar to that of the diagram 1, but contains an additional factor of the order $1/ql$

and may be omitted. Substituting the expressions for the vertices g and γ from (19) into (12) we obtain the equation for $\chi(\mathbf{q})$

$$0 = \int \frac{d^3 p d\epsilon}{(2\pi)^4} [S_0(\epsilon + \omega) - S_0(\epsilon)] \frac{dh_0(\omega)}{d\omega} \text{Im} G^A(\epsilon, \mathbf{p}) \\ \times \left[E_F^2 (\mathbf{q} \mathbf{e}_\lambda)^2 [\phi(\mathbf{q}) - \chi(\mathbf{q})] \text{Im} G^A(\epsilon + \omega, \mathbf{p} + \mathbf{q}) \right. \\ \left. + \frac{9}{\pi \nu \tau} \int \frac{d^3 p'}{(2\pi)^3} (\mathbf{p}' \mathbf{e}_\lambda)^2 [\phi(\mathbf{p}') - \chi(\mathbf{q})] \text{Im} G^A(\epsilon + \omega, \mathbf{p} + \mathbf{p}') \right]. \quad (32)$$

The integrals over p appear as follows

$$\frac{1}{\pi \nu \tau} \int \frac{d^3 p}{(2\pi)^3} \text{Im} G^A(\epsilon, \mathbf{p}) \text{Im} G^A(\epsilon + \omega, \mathbf{p} + \mathbf{q}) = \begin{cases} (1/2ql) \arctan(ql) & (q < 2p_F) \\ \sim (2p_F/q)^2 / (ql)^2 & (q > 2p_F). \end{cases} \quad (33)$$

(Compare with (44) in [4].) For $l^{-1} \ll q < 2p_F$ the integral in (33) is equal to $\pi/4ql$.

In cubic lattices the electrons do not interact directly with transverse phonons (the vertex $g_\lambda(\mathbf{q})$ for such phonons is zero). Therefore, as can be seen from equation (32), the transverse phonons in a cubic lattice are not ‘dragged’. For longitudinal phonons $e_\lambda = \mathbf{q}/q$. Thus, the following condition must hold to satisfy (32) for $ql \gg 1$

$$E_F^2 q [\phi(\mathbf{q}) - \chi(\mathbf{q})] = \frac{9}{\pi \nu \tau} \chi(\mathbf{q}) \int_0^{2p_F} \frac{d^3 p}{(2\pi)^3} \frac{(\mathbf{p} \mathbf{n})^2}{p}. \quad (34)$$

Here $\mathbf{n} = \mathbf{q}/q$. The upper limit in the integral over p is chosen in accordance with (33). The divergence of the integral at $p > 2p_F$ is purely formal, because the expression (29) for ϕ_p was derived in the linear approximation and is true only near the Fermi level. Therefore, according to (33) it is clear that the contribution of the $p > 2p_F$ domain is negligible. It is quite natural since $2p_F$ is the maximal momentum that can be transferred to an impurity by a Fermi electron without its leaving the Fermi-surface.

Finally, we obtain

$$\chi(\mathbf{q}) = \frac{ql}{ql + 24/\pi} \phi(\mathbf{q}). \quad (35)$$

From this it follows that the smaller the phonon wavevector, the more its drag is suppressed. Substituting the phonon distribution function (35) into the expression for the phonon thermal current (13) and integrating it over \mathbf{q} and ω we obtain for the relative correction to the drag thermopower β_g

$$\frac{\beta_g - \beta_g^0}{\beta_g^0} = -1.06 \frac{\hbar}{E_F \tau} \frac{u_l p_F}{T}. \quad (36)$$

To facilitate comparison with the experimental data we will rewrite this result for the drag contribution into the thermopower S_g

$$S_g = \frac{2\pi^2}{45} \frac{T^3}{n_0 e u_l^3} \left(1 - 1.06 \frac{\hbar}{E_F \tau} \frac{u_l p_F}{T} \right). \quad (37)$$

Here n_0 is the number of electrons in the unit volume. The only dependence of the drag thermopower on the impurity concentration is connected with the correction (36). Note that the factor $\hbar/E_F \tau$ in (37) has the same order as the impurity concentration $c_{imp} = N_{imp}/N$ (see (17)).

Thus, in accordance with the simple estimate (1), at low temperatures the relative correction to the drag thermopower induced by the inelastic electron scattering on impurities may be much larger than c_{imp} . Accurate study of the integral (33) shows that sufficient deviations from the first two asymptotes occur only in the narrow domain where $ql \sim 1$ where the crossover between the asymptotes takes place. Here the integral (33) quickly achieves the constant value $1/2$, and $S_g \rightarrow 0$. Hence it follows that (36) is actually valid in the whole $q_T l \gtrsim 1$ domain, and the drag thermopower is completely suppressed at $c_{imp} \sim T/\Theta_D$. Thus at low temperatures the drag effect may be suppressed even by a relatively small amount of impurities.

The result (37) was obtained in the assumption that the Umklapp processes are insufficient. However, the kinetic equation (32) can be easily generalized to incorporate these processes. To do this we must replace the arguments \mathbf{q} in the functions $\phi_{\mathbf{q}}$ and $G_{\epsilon+\omega, \mathbf{p}+\mathbf{q}}^A$ in the first term in square brackets in (32) by $\mathbf{q} + \mathbf{G}$, where \mathbf{G} is the reciprocal lattice vector, and add the summation over \mathbf{G} in this term with the restriction $\mathbf{q} + \mathbf{G} < 2p_F$, including, perhaps, the term with $\mathbf{G} = 0$ (we will omit this restriction in all formulae to make them more readable). All functions are assumed to be periodic in the reciprocal space, for example, $G^A(\epsilon, \mathbf{p} + \mathbf{G}) = G^A(\epsilon, \mathbf{p})$, $\phi_{\mathbf{p}+\mathbf{G}} = \phi_{\mathbf{p}}$, and the integral over p is taken over the first Brillouin zone. After integration over p and ϵ in (32) according to (33) we obtain

$$\chi(\mathbf{q}) \left(\sum_{\mathbf{G}} \frac{1}{|\mathbf{q} + \mathbf{G}|} + \frac{24}{\pi q^2 l} \right) = \sum_{\mathbf{G}} \frac{\phi_{\mathbf{q}+\mathbf{G}}}{|\mathbf{q} + \mathbf{G}|}. \quad (38)$$

It is interesting to investigate the effect of impurities on normal and Umklapp processes separately. In the absence of the inelastic scattering of electrons on impurities the second term in brackets in (38) is absent and the corresponding function $\chi^0(\mathbf{q})$ may be expressed as the sum of the ‘normal’ and the ‘Umklapp’ terms induced by the corresponding processes

$$\chi^0(\mathbf{q}) = \chi_N^0(\mathbf{q}) + \chi_U^0(\mathbf{q}) \quad (39)$$

where

$$\chi_N^0(\mathbf{q}) = \frac{e\tau}{m} \mathbf{qE} \quad \chi_U^0(\mathbf{q}) = \frac{e\tau}{m} \sum'_{\mathbf{G}} \frac{\mathbf{GE}}{|\mathbf{q} + \mathbf{G}|} \bigg/ \sum_{\mathbf{G}} \frac{1}{|\mathbf{q} + \mathbf{G}|}. \quad (40)$$

The prime at the sum emphasizes the obvious fact that this sum does not contain the term with $\mathbf{G} = 0$. Note that the normal term is non-zero for $q < 2p_F$ and the Umklapp term is non-zero only for those phonons which are able to induce the Umklapp processes. The total phonon distribution function has gaps on all surfaces in the phonon Brillouin zone where the Umklapp processes of a certain type are activated; these surfaces are characterized by the equations $|\mathbf{q} + \mathbf{G}| = 2p_F$ with different reciprocal lattice vectors \mathbf{G} .

Then we similarly separate the ‘normal’ and the ‘Umklapp’ contribution to $\chi(\mathbf{q})$ in (38) (the ‘normal’ contribution $\chi_N(\mathbf{q})$ is defined as $\chi(\mathbf{q})$ in the absence of the Umklapp processes, and the ‘Umklapp’ term $\chi_U(\mathbf{q})$ is the remainder). The normal term is suppressed by impurities through the same factor as in (35), and the Umklapp term is suppressed as follows (we omit the insufficient correction of the order $(Gl)^{-1}$)

$$\chi_U(\mathbf{q})/\chi_U^0(\mathbf{q}) = \sum_{\mathbf{G}} \frac{1}{|\mathbf{q} + \mathbf{G}|} \bigg/ \left(\sum_{\mathbf{G}} \frac{1}{|\mathbf{q} + \mathbf{G}|} + \frac{24}{\pi q^2 l} \right). \quad (41)$$

It is easy to see that the normal term is suppressed somewhat stronger than the Umklapp one, because the sums in (41) contain additional positive terms which effectively decrease the ratio of the impurity term to the phonon term. However, this effect is significant only for phonons with $|\mathbf{q}| \sim |\mathbf{q} + \mathbf{G}|$. At low temperatures these phonons are very rare and do not

make a sufficient contribution to the thermal current. The difference in the suppression factors for normal and Umklapp terms may be sufficient at intermediate temperatures, especially if the corresponding terms in the thermal current, being always opposite in sign, are approximately equal in magnitude.

5. Discussion

Let us compare the different kinds of phonon contributions to the thermoelectric coefficient. As we have shown above, the phonon effects in impure metals manifest themselves as corrections to the diffusive component of the thermoelectric coefficient (β_{e-ph} and $\beta_{e-ph-imp}$), as well as to the drag contribution β_g . Here we will write out the order-of-magnitude estimates for all the studied terms in the thermopower S :

$$S_d^0 \sim \frac{1}{e} \frac{T}{E_F} \quad (42a)$$

$$S_g^0 \sim \frac{1}{e} \left(\frac{T}{\Theta_D} \right)^3 \quad (42b)$$

$$S_{e-ph} \sim \frac{1}{e} \left(\frac{T}{E_F} \right)^2 \left(\frac{T}{\Theta_D} \right)^2 \frac{1}{c_{imp}} \quad (42c)$$

$$S_{e-ph-imp} \sim \frac{1}{e} \left(\frac{T}{E_F} \right)^2 \frac{T}{\Theta_D} \quad (42d)$$

$$\Delta S_g \sim -\frac{1}{e} \left(\frac{T}{\Theta_D} \right)^2 c_{imp}. \quad (42e)$$

The correction owing to the Bloch–Grüneisen contribution to conductivity is much less than the value (42c) by the parameter $(T/\Theta_D)^2$ and may be neglected, while the correction owing to interference contribution to conductivity has the same order as (42d) and is included thereto. The estimate (42c) holds if $S_{e-ph} \ll S_d^0$; this inequality coincides with the condition of the ‘weak Mattissen rule’ and may be rewritten as $c_{imp} \gg (T/E_F)(T/\Theta_D)^2$. The estimate (42e) is correct until $c_{imp} \simeq T/\Theta_D$; in the opposite case the drag thermopower is completely suppressed. Note that the impurities always decrease the magnitude of the thermopower.

Comparing different contributions (5) at a certain temperature $T \ll \Theta_D$, one can sort out three concentration domains where different corrections dominate:

$$\frac{T}{E_F} \left(\frac{T}{\Theta_D} \right)^2 \ll c_{imp} \ll \frac{T}{E_F} : S_g^0 \gg S_{e-ph} \gg \Delta S_g \quad (43a)$$

$$\frac{T}{E_F} \ll c_{imp} \ll \frac{T}{\Theta_D} : \Delta S_g \gg S_{e-ph} \gg S_{e-ph-imp} \quad (43b)$$

$$\frac{T}{\Theta_D} \ll c_{imp} \ll 1 : S_g \rightarrow 0 \quad S_{e-ph-imp} \gg S_{e-ph}. \quad (43c)$$

Thus, we see that the phonon drag effect plays the most important role in the thermopower of impure metals until it is completely suppressed at $c_{imp} \sim T/\Theta_D$. At concentrations (43b) the dependence of the thermopower on impurity concentration is determined by the suppression of the drag effect according to (42e), while at small concentrations (43a) it is governed by the Bloch–Grüneisen correction (42c). In the opposite case (43c) the interference correction (42d) can become important. In addition, in this latter region the inequality $q_T l \gg 1$ violates and the diagrams with multiple impurity lines become significant.

The obtained results are also notable for another interesting aspect. As follows from (42b), leaving the inelastic interference processes out of account, the drag thermopower S_g (which is the observed value) of an impure metal has the same magnitude as the thermopower of a very pure metal where one can neglect the electron–impurity scattering and consider only the phonon-induced electronic transport [1, 19]. This unusual feature of the thermopower is related to the fact that the decrease of the thermoelectric coefficient at the transition from pure to impure samples owing to the shortening of the electron relaxation time is accompanied by the same decrease of the conductivity. As a result, their ratio remains constant. This interesting observation highlights the role of the inelastic electron scattering on impurities in the suppression of the drag thermopower in impure metallic systems.

Experimentally, the effects studied in the present paper could be observed in dilute substitutional alloys at sufficiently low temperatures, $T \lesssim \Theta_D/5$ or even lower. We are not aware of the detailed experimental investigations of the thermopower at different concentrations in the relevant temperature range. Recently an experimental study of the thermopower was done for metastable $\text{Al}_{1-x}\text{Si}_x$ solid solutions [17] and the suppression of the drag contribution to the thermopower by small Si concentration was found to be in qualitative agreement with our conclusions. However, as the situation is complicated by a strong modification of the Al phonon spectrum in $\text{Al}_{1-x}\text{Si}_x$, the quantitative comparison with the data of [17] would be unreasonable. To verify the present theory and provide a deeper insight into the phonon effects in the thermopower of impure metals more detailed experimental studies of the different low-temperature contributions would be desirable.

In conclusion, we have studied the phonon effects in the thermopower of impure metallic systems. We found that phonons affect the diffusive component of the thermopower (the contribution of equilibrium phonons), as well as induce the drag component (the contribution of non-equilibrium phonons), and the latter one usually dominates. At the same time, the dependence of the drag thermopower on temperature and impurity concentration at low temperature is strongly affected by the processes of inelastic scattering of electrons on impurities with the emission or absorption of a phonon. These processes reduce the total magnitude of the drag thermopower and completely suppress it when the impurity concentration reaches $c_{imp} \sim T/\Theta_D$. This is the main mechanism of the low-temperature suppression of the phonon drag in impure metals and alloys.

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