Electron-ion energy exchange in simple metals in Ziman approach

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Abstract. The coefficient of the electrion-ion energy exchange in liquid aluminum is calculated within the framework of Ziman approach for electron kinetic coefficients. Calculations are made to study dependence of the electron-ion heat transfer coefficient on the electron and ion temperatures.

1. Introduction

The coefficient of heat transfer between electrons and ions in a nonequilibrium electron-ion system of a metal that occurs under the action of ultrashort laser pulses is an important kinetic coefficient, along with the coefficient of electronic heat conduction determining the dynamics of heating a target by a laser pulse [1]. At the same time, the intensity of the laser pulses can be so large that the target substance undergoes a phase transition from a solid to a liquid state [1]. An effective approach to calculating the electronic kinetic coefficients in the liquid state is Ziman approach, which uses the relaxation time approximation with allowance for the ionic structure factor for electron-ion scattering. With this approach, it is possible to obtain both single-temperature and two-temperature (at unequal temperatures of electrons and ions) values of resistivity and electronic thermal conductivity of liquid metals [2]. However, Ziman approach was not applied to the coefficient of electron-ion heat transfer. This paper shows that the Ziman approximation can also be used to calculate the energy exchange between electrons and ions in a liquid metal.

2. Ziman approach for the electron-ion energy exchange in liquid metals

The ion energy E can be represented as the sum of kinetic $E_{\rm k}$ and potential $E_{\rm p}$. Denote

$$\frac{{\rm d}E/{\rm d}t}{{\rm d}E_k/{\rm d}t} = 1 + \frac{C_{\rm vp}}{C_{\rm vk}} = \gamma(T_i).$$

Here, $C_{\rm vk}$ and $C_{\rm vp}$ are the contributions of kinetic energy and potential energy to the total isochoric heat capacity of ions C_v. Per atom $C_{\rm vk} = 3/2k_{\rm B}$ and $k_{\rm B}$ is the Boltzmann constant). Then the change in the internal energy of the ions

$$\frac{{\rm d}E}{{\rm d}t} = \gamma(T_i)\frac{{\rm d}E_k}{{\rm d}t}.$$

To calculate the rate of change of the kinetic energy of ions per unit volume, we write it in the form

$$\frac{{\rm d}E_k}{{\rm d}t} = \int \varepsilon(\mathbf{p})\frac{\partial N}{\partial t}(\mathbf{p}){\rm d}\mathbf{p}$$

Here $\partial N/\partial t(\mathbf{p}){\rm d}\mathbf{p}$ gives the rate of increase of the number of ions in the unit volume with the energy $\varepsilon(\mathbf{p}) = \mathbf{p}^2/(2M)$ in the momentum interval ${\rm d}\mathbf{p}$ and M is the mass of atom. Supposing two-temperature situation with the electron temperature T_e and ion temperature T_i we introduce the Boltzmann distribution function of ions with their concentration n_i

$$N(\mathbf{p}) = \frac{n_i}{(2\pi M k_{\rm B}T_i)^{3/2}}\exp\left(-\frac{p^2}{2Mk_{\rm B}T_i}\right)$$

and Fermi function of electrons with the energy ε' and chemical potential μ

$$f(\varepsilon') = \frac{1}{\exp(\frac{\varepsilon'-\mu}{k_{\rm B}T_i})+1}.$$

Electron states are marked with a stroke. Then considering $\mathbf{p}+\mathbf{q} \to \mathbf{p}, \mathbf{p}'-\mathbf{q} \to \mathbf{p}'$ and $\mathbf{p} \to \mathbf{p}+\mathbf{q}, \mathbf{p}' \to \mathbf{p}'-\mathbf{q}$ scattering of electron and ion with the transmitted momentum $\mathbf{q}$, we can write

$$\frac{\partial N}{\partial t}(\mathbf{p}) = \int \Phi(\mathbf{p},\mathbf{p}',\mathbf{q})W(\mathbf{p},\mathbf{p}',\mathbf{q})\frac{2V{\rm d}\mathbf{p}'}{(2\pi\hbar)^3}\frac{V{\rm d}\mathbf{q}}{(2\pi\hbar)^3}, \tag{1}$$

where $W(\mathbf{p},\mathbf{p}',\mathbf{q})$ is the probability per unit time of specified processes with the transmitted momentum $\mathbf{q}$. Using the golden rule, this probability can be presented as

$$W(\mathbf{p},\mathbf{p}',\mathbf{q}) = \frac{2\pi}{\hbar}w\delta(\alpha-\beta),$$

where $w(\mathbf{q}) = w(q)$ is the squared matrix element of electron-ion scattering with the transmitted momentum $\mathbf{q}$ and

$$\alpha = \frac{(\mathbf{p}+\mathbf{q})^2}{2M} - \frac{\mathbf{p}^2}{2M},$$
$$\beta = \frac{\mathbf{p}'^2}{2m} - \frac{(\mathbf{p}'-\mathbf{q})^2}{2m}.$$

(m is the electron effective mass). Squared matrix element $w(q)$ can be presented as

$$w(q) = \frac{S(q)}{V^2}u_q^2.$$

Here $S(q)$ is the structure factor of liquid metal and

$$u_q = \int \exp\left(-i\frac{\mathbf{qr}}{\hbar}\right)u(\mathbf{r}){\rm d}r$$

is a Fourier transform of the pseudopotential $u(\mathbf{r})$ of electron-ion interaction Statistical factor in (1)

$$\Phi(\mathbf{p},\mathbf{p}',\mathbf{q}) = N(\mathbf{p}+\mathbf{q})f(\mathbf{p}'-\mathbf{q})(1-f(\mathbf{p}')) - N(\mathbf{p})f(\mathbf{p}')(1-f(\mathbf{p}'-\mathbf{q}))$$

when introducing designations

$$\varepsilon = \frac{\mathbf{p}^2}{2M}, \qquad \varepsilon = \frac{\mathbf{p}'^2}{2m}, \qquad z = \exp\left(\frac{\varepsilon' - \mu}{k_{\rm B}T_{\rm e}}\right),$$

can be written as

$$\Phi(\alpha, \beta, \varepsilon, \varepsilon') = N(\mathbf{p})\left(\mathrm{e}^{-\alpha/k_{\rm B}T_{\rm i}} f(\varepsilon' - \beta)[1 - f(\varepsilon')] - f(\varepsilon')[1 - f(\varepsilon' - \beta)]\right)$$
$$= N(\mathbf{p})\frac{z}{z+1}\frac{\mathrm{e}^{-\alpha/k_{\rm B}T_{\rm i}} - \mathrm{e}^{-\beta/k_{\rm B}T_{\rm e}}}{z\mathrm{e}^{-\beta/k_{\rm B}T_{\rm e}} + 1}. \tag{2}$$

Then the energy transmitted from the electrons to ions per unit time and unit volume is

$$\frac{\mathrm{d}E}{\mathrm{d}t} = \int \frac{\mathbf{p}^2}{2M} N(\mathbf{p})\frac{z}{z+1}\frac{\mathrm{e}^{-\alpha/k_{\rm B}T_{\rm i}} - \mathrm{e}^{-\beta/k_{\rm B}T_{\rm e}}}{z\mathrm{e}^{-\beta/k_{\rm B}T_{\rm e}} + 1}\frac{2\pi}{\hbar} w\delta(\alpha - \beta)\frac{2V\mathrm{d}\mathbf{p}'}{(2\pi\hbar)^3}\frac{V\mathrm{d}\mathbf{q}}{(2\pi\hbar)^3}\mathrm{d}\mathbf{p} \tag{3}$$

First we integarete over $\mathbf{p}'$ in (3). Using spherical coordinates (p', θ', ϕ') with z-axes directed along $\mathbf{q}$ and denoting $\tau' = -\cos\theta'$ we obtain

$$\beta = -\frac{2p'q\tau' + q^2}{2m}$$

and after integration over ϕ'

$$\mathrm{d}\mathbf{p}' = 2\pi p'^2 \mathrm{d}p' d\tau' = -2\pi m\frac{p'}{q}\mathrm{d}p'\mathrm{d}\beta. \tag{4}$$

Analogously introducing sperical coordinates (p, θ, ϕ) for $\mathbf{p}$ with designation $\tau = -\cos\theta$, we have

$$\alpha = \frac{-2pq\tau + q^2}{2M}.$$

Then $\mathrm{d}\mathbf{p}$ can bre written as

$$\mathrm{d}\mathbf{p} = 2\pi p^2 \mathrm{d}p\mathrm{d}t = -2\pi M\frac{p}{q}\mathrm{d}\alpha\mathrm{d}p.$$

So $\mathrm{d}\mathbf{p}'\mathrm{d}\mathbf{p}$ in (3) can be written as

$$\mathrm{d}\mathbf{p}'\mathrm{d}\mathbf{p} = (2\pi)^2 Mm\frac{p'p}{q^2}\mathrm{d}p'\mathrm{d}p\mathrm{d}\alpha\mathrm{d}\beta$$

Thus, integration with respect to $\mathbf{p}'$ implies integration with respect to β. Selecting the factors depending on β in (3), we obtain at $0 \le \theta' \le \pi$ the integral over β:

$$-\int_{\frac{-2p'q-q^2}{2m}}^{\frac{2p'q-q^2}{2m}} \frac{\mathrm{e}^{-\alpha/k_{\rm B}T_{\rm i}} - \mathrm{e}^{-\beta/k_{\rm B}T_{\rm e}}}{z\mathrm{e}^{-\beta/k_{\rm B}T_{\rm e}} + 1}\delta(\beta - \alpha)d\beta, \tag{5}$$

which changes β onto α and reduces the statistical factor (2) to

$$\Phi(\alpha, \varepsilon, \varepsilon') = N(\mathbf{p})\frac{z}{z+1}\frac{\mathrm{e}^{-\alpha/k_{\rm B}T_{\rm i}} - \mathrm{e}^{-\alpha/k_{\rm B}T_{\rm e}}}{z\mathrm{e}^{-\alpha/k_{\rm B}T_{\rm e}} + 1}. \tag{6}$$

Taking into account $\alpha/k_{\rm B}T_{\rm e} << 1$ and $\alpha/k_{\rm B}T_{\rm i} << 1$ inequalities, this expression can be reduced to

$$\Phi(\alpha, \varepsilon, \varepsilon') = N(\mathbf{p})\frac{z}{(z+1)^2}\alpha\left(\frac{1}{k_{\rm B}T_{\rm e}} - \frac{1}{k_{\rm B}T_{\rm i}}\right). \tag{7}$$

To give nonzero result because of the δ-function, the inequality

$$\frac{-2p'q - q^2}{2m} \le \alpha \le \frac{2p'q - q^2}{2m} \tag{8}$$

must be carried out.

From (8)

$$\frac{-2p'q-q^2}{2m} \le \frac{-2pq\tau+q^2}{2M} \le \frac{2p'q-q^2}{2m}$$

and

$$\frac{-2p'+q}{2p\xi} \le \tau \le \frac{2p'+q}{2p\xi}. \tag{9}$$

This enequality (9) together with the restriction $-1 \le \tau \le 1$ selects the following areas of the variables p', α, p. (Here we introduced designation $\xi = m/M << 1$).

1. $p \le \frac{q}{2\xi}$, $\frac{q^2-2pq}{2M} \le \alpha \le \frac{q^2+2pq}{2M}$, $p' \ge \frac{q}{2}(1-\xi)+\frac{m\alpha}{q}$
2. $p \ge \frac{q}{2\xi}$, $\frac{q^2}{2M}(1-\frac{1}{\xi}) \le \alpha \le \frac{q^2+2pq}{2M}$, $p' \ge \frac{q}{2}(1-\xi)+\frac{m\alpha}{q}$
3. $p \ge \frac{q}{2\xi}$, $\frac{q^2-2pq}{2M}, \le \alpha \le \frac{q^2}{2M}(1-\frac{1}{\xi})$, $p' \ge \frac{q}{2}(1+\xi)-\frac{m\alpha}{q}$.

Taking into account that $\xi << 1$ these areas cam be simpler written as

1. $p \le \frac{q}{2\xi}$, $\frac{q^2-2pq}{2M} \le \alpha \le \frac{q^2+2pq}{2M}$, $p' \ge \frac{q}{2}+\frac{m\alpha}{q}$
2. $p \ge \frac{q}{2\xi}$, $-\frac{q}{2m} \le \alpha \le \frac{q^2+2pq}{2M}$, $p' \ge \frac{q}{2}+\frac{m\alpha}{q}$
3. $p \ge \frac{q}{2\xi}$, $\frac{q^2-2pq}{2M}, \le \alpha \le -\frac{q}{2m}$, $p' \ge \frac{q}{2}-\frac{m\alpha}{q}$.

With $\alpha \sim q^2/(2M) << q^2/(2m)$ contribution of small values of q is small, so the restriction on p' reduces simply to $p' \ge q/2$. Then the range of integration in the (p,α)-plane is

$$0 \ge p < \infty, \qquad \frac{q^2-2pq}{2M} \le \alpha \le \frac{q^2+2pq}{2M}, \tag{10}$$

wherein $p' \ge q/2$. Within the range (10) integration over α in (7) gives

$$\int_{\frac{q^2-2pq}{2M}}^{\frac{q^2+2pq}{2M}} \alpha \mathrm{d}\alpha = \frac{pq^3}{M^2}. \tag{11}$$

Then the right-hand side in the equality (3) reduces to a three-fold integral

$$\frac{\mathrm{d}E}{\mathrm{d}t} = \int_0^\infty \mathrm{d}q \int_{q^2/(8m)}^\infty \mathrm{d}\varepsilon' \int_0^\infty \mathrm{d}p$$
$$\frac{p^2}{2M} N(p) \frac{z(\varepsilon')}{(z(\varepsilon')+1)^2} \frac{pq^3}{M^2} \frac{2\pi}{\hbar} w \frac{2V}{(2\pi\hbar)^3} \frac{V}{(2\pi\hbar)^3} 4\pi q^2 \frac{(2\pi)^2 M m^2 p}{q^2} \left(\frac{1}{k_\mathrm{B} T_\mathrm{i}} - \frac{1}{k_\mathrm{B} T_\mathrm{e}} \right). \tag{12}$$

Calculating the integral over p, we get

$$\int_0^\infty \frac{p^2}{2M} \frac{p}{M^2} M p N(p) \mathrm{d}p = \frac{3}{8\pi} \frac{n_i k_\mathrm{B} T_\mathrm{i}}{M}.$$

Calculating the integral over ε' gives

$$\int_{q^2/(8m)}^\infty \frac{z(\varepsilon')}{(z(\varepsilon')+1)^2} \mathrm{d}\varepsilon' = \frac{k_\mathrm{B} T_\mathrm{e}}{\exp\left(\frac{q^2/(8m)-\mu}{k_\mathrm{B} T_\mathrm{e}}\right)+1}.$$

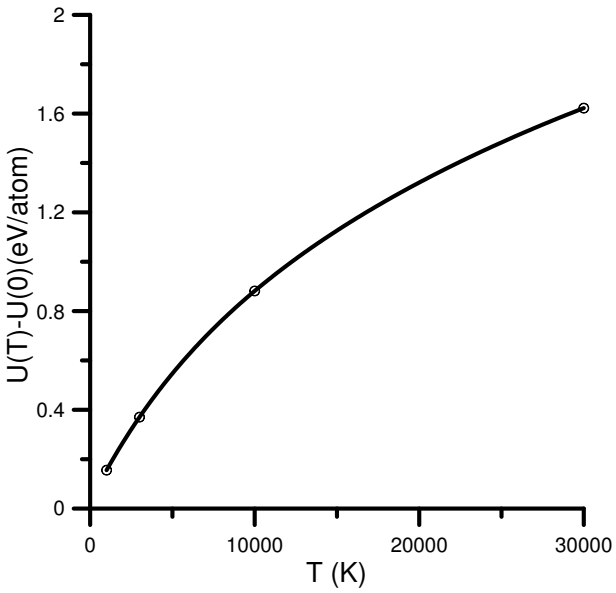

Figure 1. Change of the potential energy in liquid aluminum $U(T)-U(0)$ in dependence on the temperature. Circles are the results of the molecular dynamics modeling, solid line corresponds to the analitical fit of these results

Then the calculation of $\mathrm{d}E_k/\mathrm{d}t$ reduces to a single integration:

$$\frac{\mathrm{d}E_k}{\mathrm{d}t} = \frac{3}{8\pi}\frac{n_i}{M}\frac{2\pi}{\hbar}\frac{2V}{(2\pi\hbar)^3}\frac{V}{(2\pi\hbar)^3}(2\pi m)^2 4\pi \int_0^\infty \frac{q^3 w(q,T_e,T_i)}{\exp\left(\frac{q^2/(8m)-\mu}{k_\mathrm{B}T_\mathrm{e}}\right)+1}\mathrm{d}q \times k_\mathrm{B}(T_\mathrm{e}-T_\mathrm{i}), \tag{13}$$

giving the coefficient of electron-ion energy exchange

$$G(T_e,T_i) = \frac{3\gamma(T_i)k_B}{(2\pi)^3}\frac{n_i m^2}{M\hbar^3}\int_0^\infty \frac{k^3 S(k) u_q^2}{\exp\left(\frac{\hbar^2k^2/(8m)-\mu}{k_\mathrm{B}T_\mathrm{e}}\right)+1}\mathrm{d}k. \tag{14}$$

as a coefficient in the expression

$$\frac{\mathrm{d}E}{\mathrm{d}t} = G(T_e,T_i)(T_\mathrm{e}-T_\mathrm{i}).$$

Here $k = q/\hbar$ is a wave number.

3. Results

In figure 1 the dependence of the potential energy of atoms in liquid aluminum on the temperature is shown. Results of molecular dynamics simulations and their analitical approximatition are presented. Analitical approximation then has been differentiated with respect to temperature to give the contribution C_vp of the potential energy into the isochoric heat capacity. This enables to calculate $\gamma(T_i)$ dependence shown in figure 2. Then we take the electron-ion interaction potential as the Ashkroft potential [3] with the Fourier transform in the form, taking into account the screening of the interaction

$$u(q,x,T_\mathrm{e}) = \frac{U(q)}{\varepsilon(q,x,T_\mathrm{e})}. \tag{15}$$

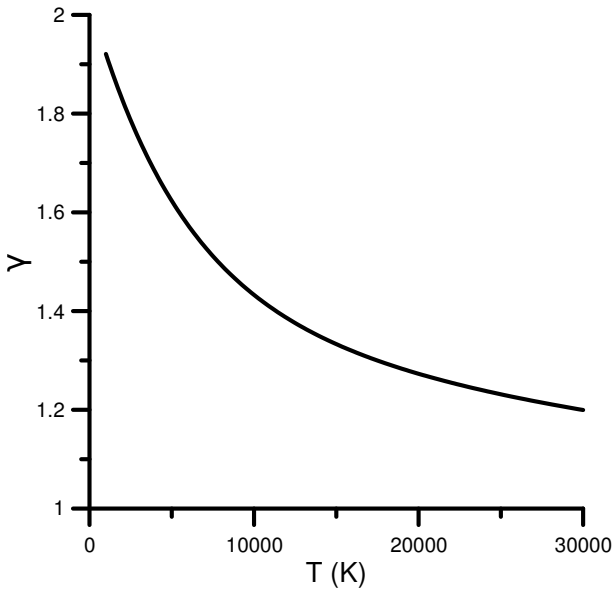

Figure 2. Dependence of the parameter γ on the ion temperature

Here $U(q)$ is the Fourier transform of Ashkroft potential $U(r)$ consisting of the empty core and Coulomn interaction outside the core:

$$U(r) = 0,\ r < r_0$$
$$U(r) = -\frac{ze^2}{r},\ r > r0.$$

Then

$$U(q) = \int U(r) e^{-i\mathbf{qr}} d\mathbf{r} = -\frac{4\pi z e^2}{q^2} \cos q\, r_0. \tag{16}$$

Dielectric function $\varepsilon(q, x, T_\mathrm{e})$ in (15), describing the electron gas screening of the Ashkroft-type interaction, was taken in the Thomas-Fermi approach:

$$\varepsilon_{\mathrm{TF}}(q) = 1 + \frac{\kappa^2(T_\mathrm{e})}{q^2}. \tag{17}$$

Here the Thomas-Fermi reverse sqreening length is

$$\kappa(T_\mathrm{e}) = \sqrt{\frac{4\pi e^2}{\partial\mu(T_\mathrm{e})/\partial n_\mathrm{e}}}.$$

with the chemical potential $\mu(T_e)$ and electron concentration n_e. Parameter r_0 was taken to be $r_0 = 1.1949$ a.u. to reproduce the experimental value of the resistivity of liquid aluminum in the melting point [2].

To find the structure factor we used the classical molecular dynamics method for modeling the motion of aluminum atoms. Interatomic potential was chosen in the framework of "embedded atom" model, taking into account many particle forces in metals. Model "embedded atom" potentials for aluminum were taken from [4, 5]. The use of the of the electron-ion interaction pseudopotential and a structure factor allows us to calculate the coefficient of electron-ion energy exchange (14). This cofficient as a function of electron temperature for values of ion temperature $T_i = 1000, 3000, 10000, 30000$ K is presented in figures 3 and 4.

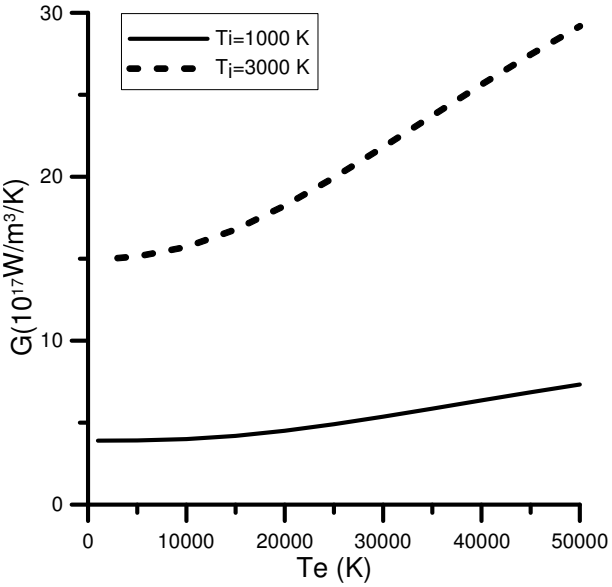

Figure 3. The coefficient of heat transfer between electrons and ions in dependence on the electron temperature T_e for two values of ion temperature $T_i = 1000$ and 3000 K.

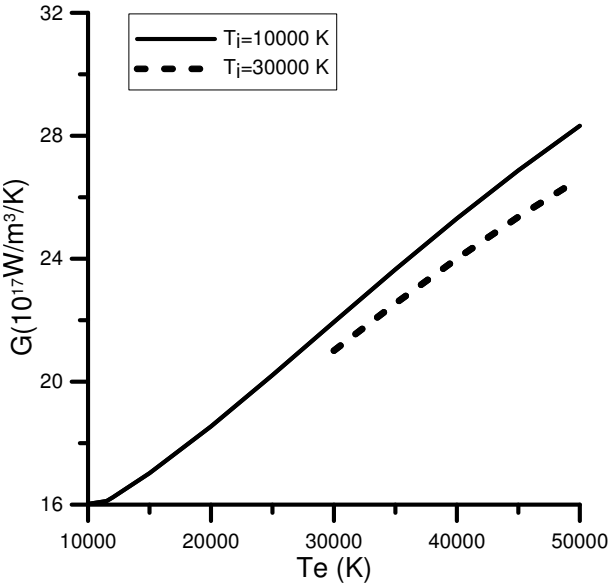

Figure 4. The coefficient of heat transfer between electrons and ions in dependence on the electron temperature T_e for two values of ion temperature $T_i = 1000$ and 3000 K.

In contrast to the electron-phonon heat transfer coefficient in solid metals, the coefficient of energy exchange between electrons and ions in the liquid metal state essentially depends not only on electron, but also ion temperature.

4. Conclusion

The coefficient of electron-ion heat transfer in a liquid metal was calculated using Ziman approach to electron transfer coefficients. Aluminum, relating to the so-called simple metals, in the spectrum of electronic excitations of which there are only s- and p-electrons, is considered.

For such a metal, the interionic interactions weakly depend on the temperature of these electrons. This made it possible to calculate the coefficient of electron-ion heat transfer depending on the electron temperature, using the structural factor calculated in a single-temperature state.

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