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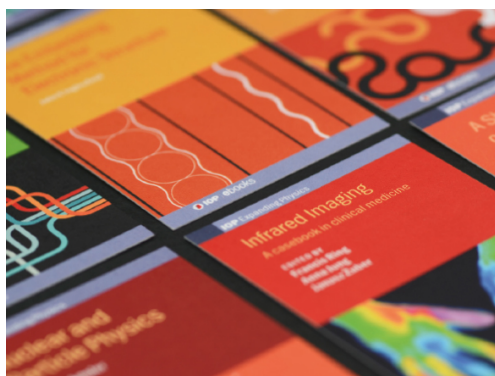
Electron–ion energy exchange in simple metals in Ziman approach

Yu V Petrov[1,2], N A Inogamov[1], K P Migdal[3], A V Mokshin[4] and B N Galimzyanov[4]

[1] Landau Institute for Theoretical Physics of the Russian Academy of Sciences, Akademika Semenova 1a, Chernogolovka, Moscow Region 142432, Russia
[2] Moscow Institute of Physics and Technology, Institutskiy Pereulok 9, Dolgoprudny, Moscow Region 141701, Russia
[3] Dukhov Automatics Research Institute (VNIIA), Federal State Unitary Enterprise, Sushchevskaya 22, Moscow 127055, Russia
[4] Kazan Federal University, Kremlyovskaya 18, Kazan, Tatarstan 420008, Russia

E-mail: `nailinogamov@gmail.com`

Abstract.   The coefficient of the electron–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 heat transfer coefficient between electrons and ions in a nonequilibrium electron–ion system of a metal, arising under the influence of ultrashort laser pulses, is an important kinetic coefficient along with the coefficient of electronic heat conduction, which determines the dynamics of target heating using a laser pulse [1, 2]. At the same time, the intensity of laser pulses can be so large that the target material undergoes a phase transition from solid to liquid [1, 3–5]. 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 [6, 7]. 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

We consider nonequlibrium two-temperature situation arising under the action of ultrashort femtosecond laser irradiation on the metal, when electrons can be characterized by their own temperature T_e different from the temperature of ions T_i [1, 8–12].

We are interesting in the rate of energy transfer from heated electrons to ions as the ion internal energy growth rate dE/dt. When the electron–ion energy exchange takes place in a

solid metal, the ion internal energy growth rate can be presented as [13]

$$\frac{\mathrm{d}E}{\mathrm{d}t} = G(T_\mathrm{e})(T_\mathrm{e} - T_\mathrm{i}), \tag{1}$$

and this linear relationship occurs not only for small differences between the electron and ion temperatures. Similar linear dependence on the difference between electron and ion temperatures is carried out at the electron–ion energy exchange in gaseous plasma [14].

Electron–ion coupling (in solid state electron–phonon coupling) is intensively investigated both experimentally [15–21] and theoretically [11, 13, 22–29] because it is an important kinetic coefficient, along with the electronic thermal conductivity coefficient determining the dynamics of heating a target by a laser pulse. For metals under consideration when we study the laser ablation, the threshold value of laser fluence for the ablation onset exceeds the melting threshold, at that the melting takes place at nonequilibrium two-temperature situation with unequal temperatures of electrons and ions. Thus it is important to calculate the electron–ion energy exchange in a liquid state of metal.

The ion internal energy E can be represented as the sum of kinetic E_k and potential E_p. We will consider a simple metal aluminum, in which the electrons of the filled valence energy bands are separated from the conduction electrons by a gap of about 70 eV and therefore are not excited at the electron temperatures under consideration. In these conditions ion internal energy slightly depends on the electron temperature and can be considered as depending on the ion temperature only [30]. Wherein we have

$$\frac{\mathrm{d}E/\mathrm{d}t}{\mathrm{d}E_\mathrm{k}/\mathrm{d}t} = \frac{C_v(T_\mathrm{i})\mathrm{d}T_\mathrm{i}/\mathrm{d}t}{C_{v\mathrm{k}}(T_\mathrm{i})\mathrm{d}T_\mathrm{i}/\mathrm{d}t} = 1 + \frac{C_{v\mathrm{p}}(T_\mathrm{i})}{C_{v\mathrm{k}}(T_\mathrm{i})} = \gamma(T_\mathrm{i}). \tag{2}$$

Here, $C_{v\mathrm{k}}$ and $C_{v\mathrm{p}}$ are the contributions of kinetic energy and potential energy to the total isochoric heat capacity of ions C_v. Kinetic energy contribution per atom is $C_{v\mathrm{k}} = 3/2k_\mathrm{B}$, where k_B is the Boltzmann constant). Then the change of the internal energy of ions $\mathrm{d}E/\mathrm{d}t$ can be obtained through the change of the kinetic energy of ions $\mathrm{d}E_\mathrm{k}/\mathrm{d}t$ and a function $\gamma(T_\mathrm{i})$:

$$\frac{\mathrm{d}E}{\mathrm{d}t} = \gamma(T_\mathrm{i})\frac{\mathrm{d}E_\mathrm{k}}{\mathrm{d}t}. \tag{3}$$

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

$$\frac{\mathrm{d}E_\mathrm{k}}{\mathrm{d}t} = \int \varepsilon(\mathbf{p})\frac{\partial N}{\partial t}(\mathbf{p})\mathrm{d}\mathbf{p}. \tag{4}$$

Here $\partial N/\partial t(\mathbf{p})\mathrm{d}\mathbf{p}$ 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 $\mathrm{d}\mathbf{p}$ 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 and the density of distribution function

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

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

$$f(\varepsilon') = \frac{1}{\exp(\frac{\varepsilon'-\mu}{k_\mathrm{B}T_\mathrm{e}}) + 1}.$$

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

$$\frac{\partial N}{\partial t}(\mathbf{p}) = \int \Phi(\mathbf{p}, \mathbf{p}', \mathbf{q})W(\mathbf{p}, \mathbf{p}', \mathbf{q})\frac{2V\mathrm{d}\mathbf{p}'}{(2\pi\hbar)^3}\frac{V\mathrm{d}\mathbf{q}}{(2\pi\hbar)^3}, \tag{5}$$

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

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

is a Fourier transform of the pseudopotential $u(\mathbf{r})$ of electron–ion interaction. Statistical factor in (5)

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

takes into account Pauli principle in the filling of the electron states [31] and when introducing designations

$$\varepsilon = \frac{\mathbf{p}^2}{2M}, \qquad \varepsilon = \frac{\mathbf{p}'^2}{2m}, \qquad z = \exp\left(\frac{\varepsilon' - \mu}{k_B T_e}\right),$$

can be written as

$$\Phi(\alpha, \beta, \varepsilon, \varepsilon') = N(\mathbf{p})\left(e^{-\alpha/(k_B T_i)}f(\varepsilon' - \beta)[1 - f(\varepsilon')] - f(\varepsilon')[1 - f(\varepsilon' - \beta)]\right)$$

$$= N(\mathbf{p})\frac{z}{z+1}\frac{e^{-\alpha/(k_B T_i)} - e^{-\beta/(k_B T_e)}}{ze^{-\beta/(k_B T_e)} + 1}. \tag{6}$$

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

$$\frac{\mathrm{d}E_k}{\mathrm{d}t} = \int \frac{\mathbf{p}^2}{2M}\frac{N(\mathbf{p})z}{z+1}\frac{e^{-\alpha/(k_B T_i)} - e^{-\beta/(k_B T_e)}}{ze^{-\beta/(k_B T_e)} + 1}\frac{2\pi}{\hbar}w\delta(\alpha - \beta)\frac{2V\mathrm{d}\mathbf{p}'}{(2\pi\hbar)^3}\frac{V\mathrm{d}\mathbf{q}}{(2\pi\hbar)^3}\mathrm{d}\mathbf{p}. \tag{7}$$

First we integarete over $\mathbf{p}'$ in (7). 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',

$$\mathrm{d}\mathbf{p}' = 2\pi p'^2 \mathrm{d}p' d\tau' = -2\pi m \frac{p'}{q}\mathrm{d}p'\mathrm{d}\beta. \tag{8}$$

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 $d\mathbf{p}$ can be written as

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

So $d\mathbf{p}'d\mathbf{p}$ in (7) can be written as

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

Thus, integration with respect to $\mathbf{p}'$ implies integration with respect to β. Selecting the factors depending on β in (7), we obtain at $0 \leqslant \theta' \leqslant \pi$ the integral over β:

$$-\int_{\frac{-2p'q-q^2}{2m}}^{\frac{2p'q-q^2}{2m}} \frac{e^{-\alpha/(k_B T_i)} - e^{-\beta/(k_B T_e)}}{z e^{-\beta/(k_B T_e)} + 1} \delta(\beta - \alpha) d\beta, \tag{9}$$

which changes β onto α and reduces the statistical factor (6) to

$$\Phi(\alpha, \varepsilon, \varepsilon') = N(\mathbf{p}) \frac{z}{z+1} \frac{e^{-\alpha/(k_B T_i)} - e^{-\alpha/(k_B T_e)}}{z e^{-\alpha/(k_B T_e)} + 1}. \tag{10}$$

Taking into account $\alpha/(k_B T_e) \ll 1$ and $\alpha/(k_B T_i) \ll 1$ inequalities ($\alpha/(k_B T_i) < \varepsilon_F m/(M k_B T_i) \ll 1$ at the ion temperatures under consideration not smaller than the melting temperature), this expression can be reduced to

$$\Phi(\alpha, \varepsilon, \varepsilon') = N(\mathbf{p}) \frac{z}{(z+1)^2} \alpha \left(\frac{1}{k_B T_e} - \frac{1}{k_B T_i} \right). \tag{11}$$

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

$$\frac{-2p'q - q^2}{2m} \leqslant \alpha \leqslant \frac{2p'q - q^2}{2m} \tag{12}$$

must be carried out. From (12) we obtain

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

and

$$\frac{-2p' + q}{2p\xi} \leqslant \tau \leqslant \frac{2p' + q}{2p\xi}. \tag{13}$$

The enequality (13) together with the restriction $-1 \leqslant \tau \leqslant 1$ selects the following areas of the variables p', α, p (here we introduced designation $\xi = m/M \ll 1$):

$$p \leqslant \frac{q}{2\xi}, \qquad \frac{q^2 - 2pq}{2M} \leqslant \alpha \leqslant \frac{q^2 + 2pq}{2M}, \qquad p' \geqslant \frac{q}{2}(1-\xi) + \frac{m\alpha}{q};$$

$$p \geqslant \frac{q}{2\xi}, \qquad \frac{q^2}{2M}(1 - \frac{1}{\xi}) \leqslant \alpha \leqslant \frac{q^2 + 2pq}{2M}, \qquad p' \geqslant \frac{q}{2}(1-\xi) + \frac{m\alpha}{q};$$

$$p \geqslant \frac{q}{2\xi}, \qquad \frac{q^2 - 2pq}{2M} \leqslant \alpha \leqslant \frac{q^2}{2M}(1 - \frac{1}{\xi}), \qquad p' \geqslant \frac{q}{2}(1+\xi) - \frac{m\alpha}{q}.$$

Taking into account that $\xi \ll 1$ these areas can be simpler written as

$$p \leqslant \frac{q}{2\xi}, \qquad \frac{q^2 - 2pq}{2M} \leqslant \alpha \leqslant \frac{q^2 + 2pq}{2M}, \qquad p' \geqslant \frac{q}{2} + \frac{m\alpha}{q};$$

$$p \geqslant \frac{q}{2\xi}, \qquad -\frac{q}{2m} \leqslant \alpha \leqslant \frac{q^2 + 2pq}{2M}, \qquad p' \geqslant \frac{q}{2} + \frac{m\alpha}{q};$$

$$p \geqslant \frac{q}{2\xi} \qquad \frac{q^2 - 2pq}{2M} \leqslant \alpha \leqslant -\frac{q}{2m}, \qquad p' \geqslant \frac{q}{2} - \frac{m\alpha}{q}.$$

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

$$0 \leqslant p < \infty, \qquad \frac{q^2 - 2pq}{2M} \leqslant \alpha \leqslant \frac{q^2 + 2pq}{2M}, \tag{14}$$

wherein $p' \geqslant q/2$. Within the range (14) integration over α in (11) gives

$$\int_{\frac{q^2-2pq}{2M}}^{\frac{q^2+2pq}{2M}} \alpha \mathrm{d}\alpha = \frac{pq^3}{M^2}. \tag{15}$$

Then the right-hand side in the equality (7) reduces to a three-fold integral

$$\frac{\mathrm{d}E}{\mathrm{d}t} = \int_0^\infty \mathrm{d}q \int_{q^2/(8m)}^\infty \mathrm{d}\varepsilon' \int_0^\infty \mathrm{d}p \frac{p^2}{2M} N(p) \frac{z(\varepsilon')}{(z(\varepsilon')+1)^2} \frac{pq^3}{M^2} \frac{2\pi}{\hbar} w \frac{2V}{(2\pi\hbar)^3} \frac{V}{(2\pi\hbar)^3} 4\pi q^2 \frac{(2\pi)^2 M m^2 p}{q^2}$$
$$\times \left(\frac{1}{k_\mathrm{B} T_\mathrm{i}} - \frac{1}{k_\mathrm{B} T_\mathrm{e}} \right). \tag{16}$$

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

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

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

$$\frac{\mathrm{d}E_\mathrm{k}}{\mathrm{d}t} = \frac{3}{8\pi} \frac{n_i}{M} \frac{2\pi}{\hbar} \frac{2V}{(2\pi\hbar)^3} \frac{V}{(2\pi\hbar)^3} (2\pi m)^2 4\pi k_\mathrm{B} (T_\mathrm{e} - T_\mathrm{i}) \int_0^\infty \frac{q^3 w(q, T_\mathrm{e}, T_\mathrm{i})}{\exp\left(\frac{q^2/(8m)-\mu}{k_\mathrm{B} T_\mathrm{e}}\right) + 1} \mathrm{d}q. \tag{17}$$

With the use of (3) it gives

$$G(T_\mathrm{e}, T_\mathrm{i}) = \frac{3\gamma(T_\mathrm{i}) k_\mathrm{B}}{(2\pi)^3} \frac{n_i m^2}{M \hbar^3} \int_0^\infty \frac{k^3 S(k) u_q^2}{\exp\left(\frac{\hbar^2 k^2/(8m)-\mu}{k_\mathrm{B} T_\mathrm{e}}\right) + 1} \mathrm{d}k \tag{18}$$

as a coefficient in the expression

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

Here $k = q/\hbar$ is a wave number. As seen from (18), unlike the case of electron–ion heat transfer in solid metals, the electron–ion coupling in liquids depends on both electron and ion temperatures.

3. Structure factor, electron–ion pseudopotential

So to calculate the electron–ion coupling we need to know the structure factor $S(k)$ and $\gamma(T_\mathrm{i})$ parameter that characterizes the relative contribution of potential and kinetic energies into the internal energy of ions. Both quantities has been calculated by using 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" potential for aluminum was taken from [32, 33].

The structure factor $S(k, T, n_i)$ in the equilibrium one-temperature case was calculated within the molecular dynamics approach with the number of aluminum atoms taken to be 13 500 and 48688. With such a change in the number of particles, the results were close. The time step for
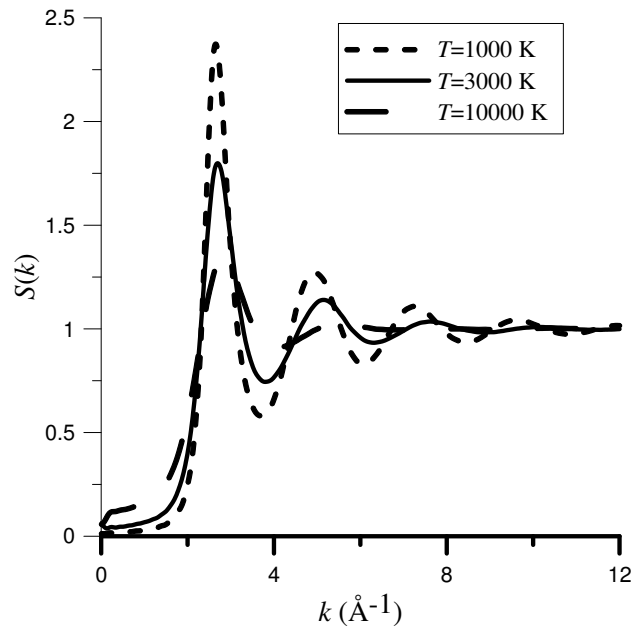
Figure 1. The structure factor of liquid aluminum at the temperature $T = 1, 3, 10$ kK and the density 2.35 g/cm^3.
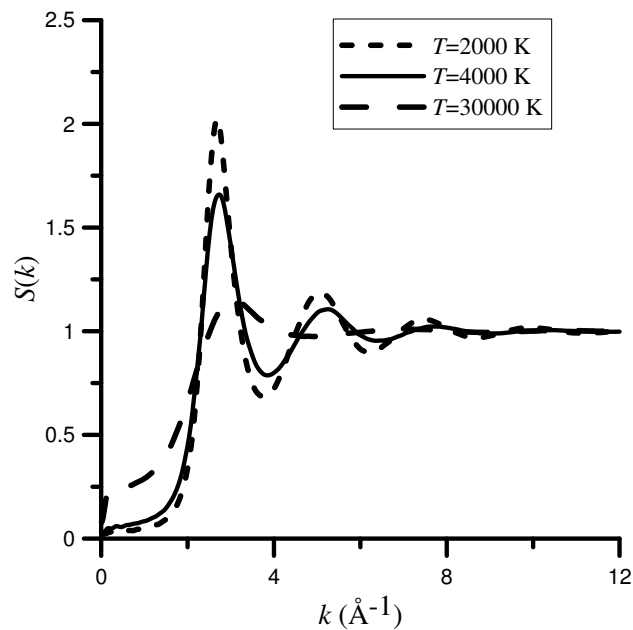
Figure 2. The structure factor of liquid aluminum at the temperature $T = 2, 4, 30$ kK. The density is equal to 2.35 g/cm^3.

the ion motion was taken to be 1 fs. We have done our calculations for liquid aluminum with a density of 2.35 g/cm^3. The structure factor for the temperature $T = 1, 3, 10$ kK is shown in figure 1 and for the temperature $T = 2, 4, 30$ kK—in figure 2.

Other value needed to calculate the electron–ion coupling parameter is the pseudopotential of the electron–ion interaction. We take the electron–ion interaction potential as the Ashcroft
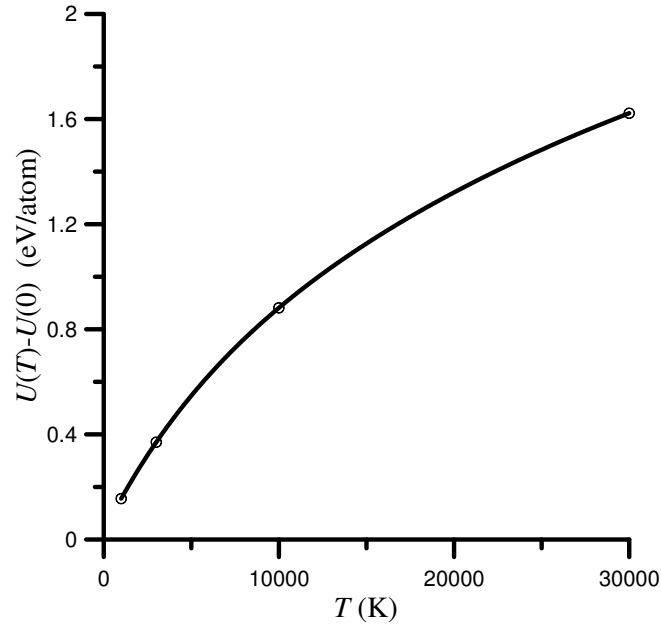
Figure 3. Change of the potential energy in liquid aluminum $U(T) - U(0)$ in dependence on the temperature. Circles are the results of the molecular dynamics modeling, solid line corresponds to the analytical fit of these results.

potential [34] with the Fourier transform in the form, taking into account the screening of the interaction

$$u(q, x, T_{\mathrm{e}}) = \frac{U(q)}{\varepsilon(q, x, T_{\mathrm{e}})}. \tag{19}$$

Here $U(q)$ is the Fourier transform of Ashcroft potential $U(r)$ consisting of the empty core of the radius r_0 and Coulomb interaction outside the core:

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

Then

$$U(q) = \int U(r) e^{-i\mathbf{q}\mathbf{r}} d\mathbf{r} = -\frac{4\pi ze^2}{q^2} \cos q \, r_0. \tag{20}$$

Dielectric function $\varepsilon(q, x, T_{\mathrm{e}})$ in (19), describing the electron gas screening of the Ashcroft-type interaction, was taken in the Thomas–Fermi approach:
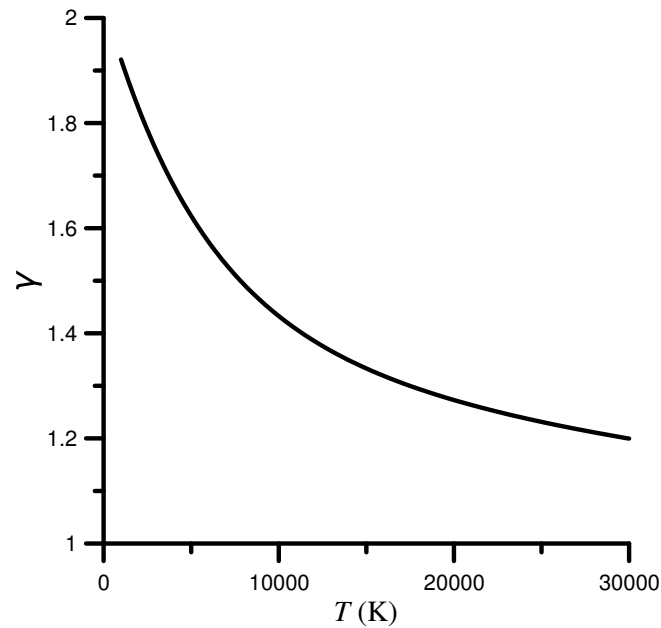
$$\varepsilon_{\mathrm{TF}}(q) = 1 + \frac{\kappa^2(T_{\mathrm{e}})}{q^2}. \tag{21}$$
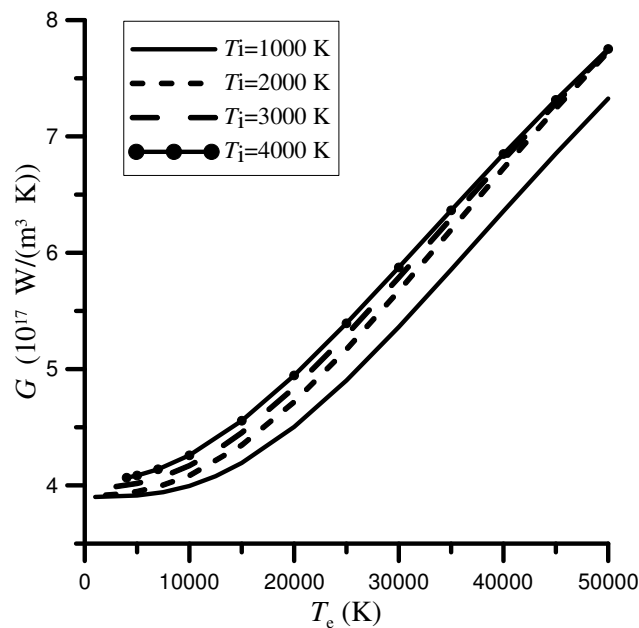
Here the Thomas–Fermi reverse screening length is

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

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 [7].

Figure 4. Dependence of the parameter γ on the ion temperature.

Figure 5. The coefficient of heat transfer between electrons and ions in dependence on the electron temperature T_e for the values of ion temperature $T_\mathrm{i} = 1000$, 2000, 3000 and 4000 K. The density is equal to 2.35 g/cm^3.

4. Results
Figure 3 shows the temperature dependence of the potential energy of atoms in liquid aluminum. The results of molecular dynamics modeling and their analytical approximations are presented. The analytical approximation is then differentiated by temperature in order to contribute $C_{v\mathrm{p}}$ of potential energy to the isochoric heat capacity. This allows us to calculate the dependence $\gamma(T_\mathrm{i})$, shown in figure 4.
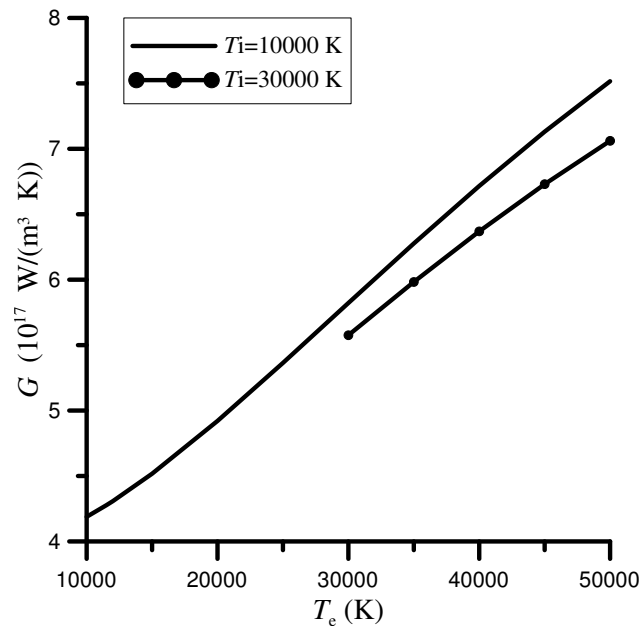
Figure 6. The electron–ion coupling in a liquid aluminum of the density of to 2.35 g/cm^3 in dependence on the electron temperature T_e for two values of ion temperature $T_i = 10\,000$ and $30\,000$ K.

The use of the electron–ion interaction pseudopotential and a structure factor allows us to calculate the coefficient of electron–ion energy exchange (18). This coefficient as a function of electron temperature for values of ion temperature $T_i = 1000, 2000, 3000, 4000$ K and $T_i = 10\,000, 30\,000$ K is presented respectively in figures 5 and 6.

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 depends not only on electron, but also ion temperature. The dependence on the ion temperature is not too significant.

5. Conclusion

The electron–ion heat transfer coefficient in liquid metal was calculated within the framework of the 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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