# Two-temperature kinetic coefficients of liquid aluminum in the conditions arising under the action of a femtosecond laser pulse

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# Two-temperature kinetic coefficients of liquid aluminum in the conditions arising under the action of a femtosecond laser pulse

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**Abstract.** Resistivity and thermal conductivity coefficient are calculated in the framework of Ziman approach. Resistivity is calculated as a reverse conductivity with the use of the relaxation time approximation accompanied by its calculation using Ziman–Evans formula. Both resistivity and thermal conductivity and Lorenz number are calculated as in the equilibrium one-temperature state and the two-temperature situation with different electron and ion temperatures. Influence of different approaches to the screening of electron–ion interactions when considering the kinetic coefficients is investigated.

## 1. Introduction

Kinetic coefficients of metals are of great importance for modeling the process of their ablation under the action of ultrashort laser pulses. When the intensity of the laser irradiation is sufficient to melt the metal, it is necessary to know these kinetic coefficients in the liquid phase in addition to their values in the solid state. Effective method of the calculation of kinetic coefficients in the liquid state of metal is the Ziman approach [1] which uses the electron–ion interaction potential and the ion structure factor as the components of the calculation. We have calculated the structure factor by the use of the molecular dynamics method in its classical version to describe the ion motion with previously obtained interatomic potentials. Such approach is less expensive in terms of spent computing resources than the use of quantum molecular dynamics approach with the subsequent application of Kubo–Greenwood method for the kinetic coefficients. The second key value in the Ziman approach is the electron–ion interaction potential, determining the value of the frequency of the electron–ion collisions and its contribution into the kinetic processes. The value of the electron–ion interaction depends upon the dielectric function of the electron gas in a metal which defines the screening of the electron–ion interactions.

We consider the influence of the choice of dielectric function in the form of Lindhard, Thomas–Fermi [2] and Hubbard [3] functions onto the results of calculation of resistivity and thermal

conductivity coefficient of liquid Al. The characteristic feature of the interaction of ultrashort laser pulses with metals is the occurrence of two-temperature states in them with the temperature of electrons exceeding the temperature of ions in the early stage of interaction, and the kinetic coefficients of a liquid metal at this stage are of great importance. We have calculated the two-temperature resistivity and the two-temperature thermal conductivity of liquid aluminum in the relaxation time approach.

## 2. The structure factor

Of the quantities, characteristic for the Ziman approach to the calculation of kinetic coefficients of liquid metals, the structure factor in a liquid state is the important component. We have calculated the structure factor $S(q)$ of a liquid aluminum through the calculation of the radial pair distribution function $g(r)$ and the use of the relation

$$S(q) = 1 + n_{\mathrm{i}} \int_0^\infty (g(r) - 1) \frac{\sin qr}{qr} 4\pi r^2 \mathrm{d}r. \tag{1}$$

To find the radial pair distribution function we used the classical molecular dynamics method for the description of the motion of aluminum atoms [4]. Interatomic potential was chosen in the model of "embedded atom" [5]. It takes into account many-particle character of interatomic forces in metals. Potentials of the embedded atom model for aluminum were taken from [6, 7] and from the other side as in [8]. The potential function derived in [8] was extensively checked in the molecular dynamics simulation of the ablation processes in aluminum with the formation of nanostructures on its surface [9,10] and in investigation of excitation of shock waves produced by the ultrashort laser pulses in the aluminum target [11]. Results for the pair distribution function with the number of atoms reaching up to 50 000, and as a consequence for the structural factor, are close in both cases of potentials [6,7] and [8]. The structure factor of liquid aluminum at the density 2.35 g/cm$^3$ is shown in figure 1 for the temperatures $T = 1$, 2 and 4 kK. It is calculated with the use of the interatomic potential for the aluminum obtained in [8].

For the temperatures $T = 3$, 10, 30 kK the structure factor calculated with the potentials [6, 7] was shown in [12]. This is the equilibrium situation, when the temperatures of ions and electrons are equal. In the aluminum, as a simple metal, conduction s- and p-electrons are strongly separated in the energy scale from electrons of inner shells (a gap is about 70 eV). Thus interatomic interaction in aluminum weakly depends on the electron temperature [13, 14] in the electron temperature range under consideration up to 60 kK in contrast to the transition metals, for example noble metals such as copper, silver, gold with their d-electrons, relatively easy excited with the electron temperature increase. Therefore the structure factor obtained in the equilibrium one-temperature state can be used also in the case of the two-temperature situation when the temperature of electrons is higher than the temperature of ions, and this temperature excess can be very significant, reaching several eV. This fact will be used in the calculation of kinetic coefficients such as the resistivity and thermal conductivity in the two-temperature state.

## 3. Electron–ion interaction and its screening

An important part of Ziman approach [1] is the electron–ion interaction giving the main contribution into the electron relaxation in the electron transfer processes. Ziman-type formulas for the kinetic coefficients consist of the form-factor, the Fourier transform of the electron–ion interaction potential besides of the ionic structure factor. We take the electron–ion interaction potential as the Ashcroft potential [15] with the Fourier transform in the form, taking into account the screening of the interaction

$$u(q, x, T_{\mathrm{e}}) = \frac{U(q)}{\varepsilon(q, x, T_{\mathrm{e}})}. \tag{2}$$

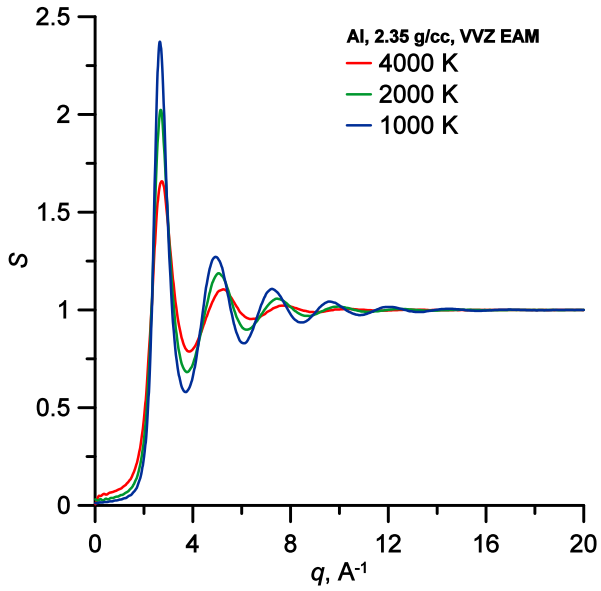


**Figure 1.** The structure factor $S(q)$ of liquid Al at the equilibrium one-temperature state for the density 2.35 g/cm$^3$. Wave number $q$ is measured in units of reverse Ångström. Temperatures are $T = 1$, 2, 4 kK. Calculations are made with the use of interatomic potentials derived in [8] (VVZ).

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

$$U(r) = \begin{cases} 0 & \text{for } r \leqslant r_0, \\ -\dfrac{ze^2}{r} & \text{for } r > r_0 \end{cases} \tag{3}$$

and

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

The number of conduction electrons per atom $z$ is chosen to be 3 because we consider the density of aluminum 2.35 g/cm$^3$. This density is far from the supposed density of the metal–dielectric transition, which is estimated to be less than 1.8 g/cm$^3$ [16, 17]. Dielectric function $\varepsilon(q, x, T_\mathrm{e})$ in (2) describes the electron gas screening of the Ashcroft-type interaction. In this work we analyzed the influence of the choice of the dielectric function onto the results of calculation of kinetic coefficients.

We have considered the Lindhard dielectric function [2, 18]

$$\varepsilon_\mathrm{L}(q) = 1 + \frac{4}{\pi a_\mathrm{B}\, q^3}\int_0^\infty k \ln\left|\frac{k+q/2}{k-q/2}\right| f(k, x, T_\mathrm{e})\,\mathrm{d}k, \tag{5}$$

where $a_\mathrm{B}$ is the Bohr radius, $f(k, x, T_\mathrm{e})$ is the Fermi distribution function, giving the dependence of the Lindhard permittivity on the electron temperature. In the long wave limit at wave numbers $q$, much smaller than the Fermi wave number $k_\mathrm{F}$, the expression (5) goes to the temperature dependent Thomas–Fermi permittivity [2]

$$\varepsilon_\mathrm{TF}(q) = 1 + \frac{4\pi e^2}{q^2}\frac{\partial n_\mathrm{e}}{\partial \mu} = 1 + \frac{\kappa^2(T_e)}{q^2}. \tag{6}$$

Here the Thomas–Fermi reverse screening length

$$\kappa(T_{\rm e}) = \sqrt{\frac{4\pi e^2}{\partial\mu(T_{\rm e})/\partial n_{\rm e}}} \tag{7}$$

depends on the electron density through the chemical potential.

Lindhard and Thomas–Fermi permittivity are considered in Hartree approach. To include exchange and correlation effects we used the Hubbard dielectric function [3]

$$\varepsilon_{\rm H}(q) = \frac{1+(1-G_{\rm H}(q))(\varepsilon_j(q)-1)}{1-G_{\rm H}(q)(\varepsilon_j(q)-1)}, \tag{8}$$

where $\varepsilon_j = \varepsilon_{\rm L}$ or $\varepsilon_{\rm TF}$ and Hubbard correction function is

$$G_{\rm H}(q) = \frac{1}{2}\frac{q^2}{q^2+k_{\rm F}^2}. \tag{9}$$

## 4. Resistivity. Two-temperature state approach

Original Ziman approach is used to obtain directly the resistivity. Introducing the function

$$U_S(k) = \int_0^{2k} q^3 S(q,T,n_{\rm i})|u(q)|^2 {\rm d}q, \tag{10}$$

Ziman formula for the resistivity can be written as

$$\rho(T,n_{\rm i}) = \frac{1}{\sigma} = \frac{3\pi}{4}\frac{m^2 n_{\rm i}}{e^2\hbar^3 k_{\rm F}^6} U_S(k_{\rm F}). \tag{11}$$

Integration over the transmitted wave number $q$ is here restricted by the $2k_{\rm F}$ in accordance with the temperature $T$, small with respect to the Fermi temperature $T_{\rm F}$. Strong excitation of electrons under the more high temperatures with the significant deviation of the Fermi distribution function from the zero temperature step function can be taken into account in the framework of the Ziman–Evans formula [19] also directly for the resistivity. In the two-temperature state with different temperatures of ions $T_{\rm i}$ and electrons $T_{\rm e}$ Ziman–Evans formula for the resistivity has a form

$$\begin{aligned}\rho(T_{\rm e},T_{\rm i},n_{\rm i}) &= \frac{4}{3\pi}\frac{m^2e^2}{\hbar^3 n_{\rm i}(4\pi z e^2)^2}\int_0^\infty \left(-\frac{\partial f}{\partial k}\right) U_S(k){\rm d}k \\ &= \frac{4}{3\pi}\frac{m^2e^2}{\hbar^3 n_{\rm i}(4\pi z e^2)^2}\int_0^\infty f\left(\frac{k}{2},T_{\rm e}\right) k^3 S(k,T_{\rm i},n_{\rm i})|u(k)|^2{\rm d}k.\end{aligned} \tag{12}$$

At $T_{\rm e} \ll T_{\rm F}$ (12) is converted to the Ziman formula for the resistivity. From the other hand, resistivity can be obtained as the value, reverse to the conductivity. To calculate the conductivity we can use ordinary for the relaxation time approach expression [20]

$$\sigma = \frac{2}{3}e^2\frac{4\pi}{(2\pi)^3}\int_0^\infty \tau(k)\left(-\frac{\partial f}{\partial\varepsilon_k}\right)\left(\frac{\hbar k}{m}\right)^2 k^2{\rm d}k. \tag{13}$$

with the electron relaxation time $\tau(k)$ being dependent upon the electron wave number $k$. In (13) $\varepsilon_k$ is the energy of electron with the wave number $k$. Within the effective mass approach we suppose $\varepsilon_k = \hbar^2k^2/(2m_s)$ with the effective mass of s and p-electrons $m_s$ to be 1.06 of free electron mass. Introducing the structure factor $S(q,T_{\rm i},T_{\rm e},n_{\rm i}) = S(q,T_{\rm i},n_{\rm i})$ with taking into account its weak dependence upon the electron temperature for aluminum and using the Born approach for the electron–ion interaction, we can write the relaxation time as

$$\tau(k,T_{\rm i}) = 4\pi\frac{\hbar^3}{mn_{\rm i}}\frac{k^3}{U_S(k)}. \tag{14}$$

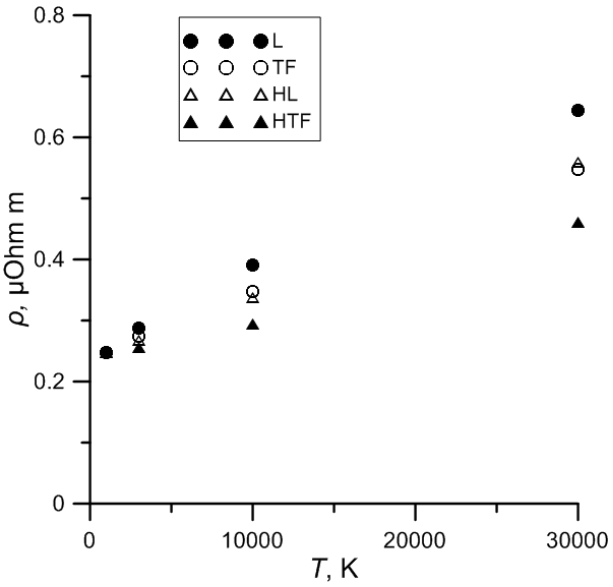


**Figure 2.** Resistivity of liquid Al in the Ziman–Evans approach at equilibrium one-temperature state depending on the temperature at different types of the screening of the electron–ion interactions. Melt density is equal to 2.35 g/cm$^3$. L—Lindhard; TF—Thomas–Fermi; HL—Hubbard with $\varepsilon_j = \varepsilon_{\rm L}$; HTF—Hubbard with $\varepsilon_j = \varepsilon_{\rm TF}$.

Then the conductivity (13) in the two-temperature state is [12]

$$\sigma(T_{\rm e}, T_{\rm i}, n_{\rm i}) = \frac{4}{3\pi}\frac{\hbar^5 e^2}{m^3 n_{\rm i}}\int_0^\infty \frac{k^7}{U_S(k)}\left(-\frac{\partial f}{\partial \varepsilon_k}(T_{\rm e})\right){\rm d}k. \tag{15}$$

And the two-temperature resistivity now can be obtained as the reverse to this value:

$$\rho(T_{\rm e}, T_{\rm i}, n_{\rm i}) = 1/\sigma(T_{\rm e}, T_{\rm i}, n_{\rm i}). \tag{16}$$

Resistivity obtained for different types of the electron gas dielectric functions is shown in the Ziman–Evans approach in figure 2 and in the reverse conductivity approach in figure 3. Use of Lindhard and Thomas–Fermi dielectric functions gives not very different results, close to those obtained in the framework of Kubo–Greenwood approach and quantum molecular dynamics in [21]. When using Ziman–Evans approach, resistivity is higher than when using the inverse conductivity approach.

## 5. Electron thermal conductivity. Two-temperature model

The main purpose of our investigations is the calculation of the electron thermal conductivity of liquid aluminum especially in its two-temperature state while the resistivity has been studied to test the approach used. Relaxation time (14) for the electron–ion interaction can be used to find two-temperature electron thermal conductivity coefficient. It can be expressed through the Onsager coefficients. Within the relaxation time approach Onsager coefficients in the two-temperature state are [22]

$$I_n(T_{\rm e}, T_{\rm i}, n_{\rm i}) = \frac{\hbar^2}{3\pi^2 m_s^2}\int k^4 \tau(k)\left(-\frac{\partial f_s}{\partial \varepsilon_k}\right)(\varepsilon_k - \mu)^n {\rm d}k, \tag{17}$$

$n = 0,\ 1,\ 2$. In the absence of the electrical current the thermal conductivity coefficient can be obtained as

$$\kappa_{\rm ei}(T_{\rm e}, T_{\rm i}, n_{\rm i}) = \frac{1}{T_{\rm e}}\left(I_2 - \frac{I_1^2}{I_0}\right). \tag{18}$$

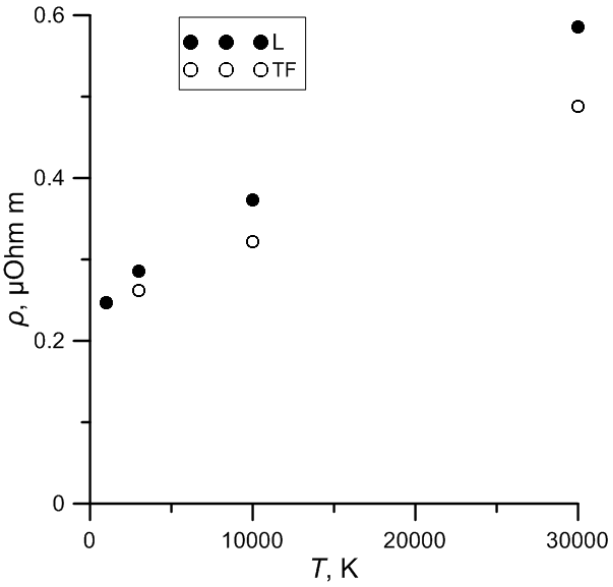


**Figure 3.** Resistivity of liquid aluminum when using the reverse conductivity approach at the one-temperature state in dependence on the temperature at two types of the electron dielectric function. Density is equal to 2.35 g/cm$^3$. L—Lindhard; TF—Thomas–Fermi.

Of them the coefficient $I_0$ is simply $I_0 = \sigma(T_\mathrm{e}, T_\mathrm{i}, x)/e^2$.

In our investigation we consider electron temperatures up to several eV arising in metals under the action of femtosecond laser pulses. This growth of the electron temperature leads to a strong increase of electron–electron interactions in a metal which contribute to the electron relaxation at the electron thermal conductivity [12]. The electron thermal conductivity coefficient when taking into account both the electron–ion and electron–electron scattering processes can be expressed through the partial coefficients [12] $\kappa_\mathrm{ei}(T_\mathrm{e}, T_\mathrm{i}, n_\mathrm{i})$ and $\kappa_\mathrm{ee}(T_\mathrm{e}, n_\mathrm{i})$ as

$$\kappa(T_\mathrm{e}, T_\mathrm{i}, n_\mathrm{i}) = \left(\kappa_\mathrm{ei}(T_\mathrm{e}, T_\mathrm{i}, n_\mathrm{i})^{-1} + \kappa_\mathrm{ee}(T_\mathrm{e}, n_\mathrm{i})^{-1}\right)^{-1}. \tag{19}$$

Electron thermal conductivity due to the electron–electron scattering $\kappa_\mathrm{ee}(T_\mathrm{e}, n_\mathrm{i})$ is taken as in [12].

In figure 4 the electron thermal conductivity is shown with only electron–ion scattering taking into account and with allowing for the electron–electron scattering in the one-temperature state. At large temperatures electron–electron scattering significantly restricts the thermal conductivity growth. Figure 5 presents results of the calculations of the thermal conductivity in the two-temperature state in dependence on the electron temperature for two ion temperatures—$T_\mathrm{i}$ = 1000 and 3000 K. For greater ion temperatures, $T_\mathrm{i}$ = 10 000 and 30 000 K, similar dependence is shown in figure 6.

## 6. Two-temperature Wiedemann–Franz ratio

Knowing the electron thermal conductivity and resistivity, we can now calculate the Lorenz number both in the equilibrium state being characterized by the temperature $T$, common for electrons and ions,

$$L(x, T) = \frac{\kappa(x, T)\rho(x, T)}{T} \tag{20}$$

and the Lorenz number

$$L_\mathrm{e}(x, T_\mathrm{e}, T_\mathrm{i}) = \frac{\kappa(x, T_\mathrm{e}, T_\mathrm{i})\rho(x, T_\mathrm{e}, T_\mathrm{i})}{T_\mathrm{e}}, \tag{21}$$

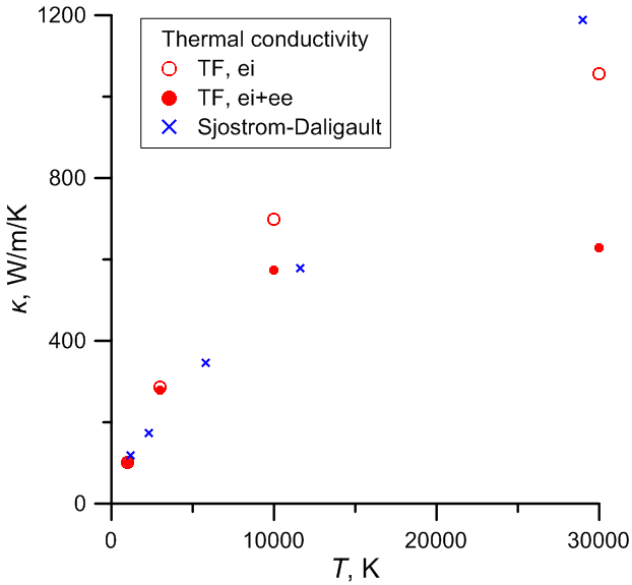


**Figure 4.** Electron thermal conductivity of liquid Al at the equilibrium one-temperature state depending on the temperature. Melt density is equal to 2.35 g/cm$^3$. Thermal conductivity coefficient calculated with only the electron–ion scattering taken into account is marked by empty circles. Results obtained when including in addition the electron–electron scattering are shown by the full circles. Also results of quantum molecular dynamics calculations with the Kubo–Greenwood approach [23] are shown.

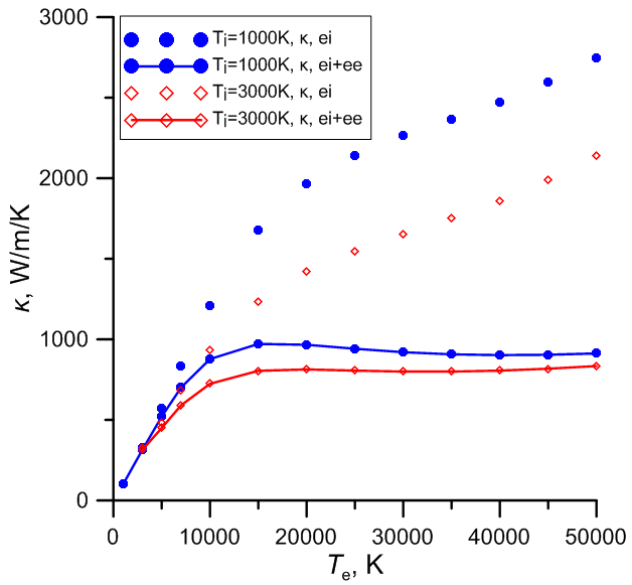


**Figure 5.** Electron thermal conductivity of liquid Al at 2.35 g/cm$^3$ in the two-temperature state in dependence on the electron temperature. Thermal conductivity coefficient is calculated with only the electron–ion scattering (ei) and with the electron–electron scattering taken into account (ei+ee).

in our nonequilibrium two-temperature situation, which is a function of the temperature of both ions and electrons separately. The equilibrium Lorentz number as a function of temperature is

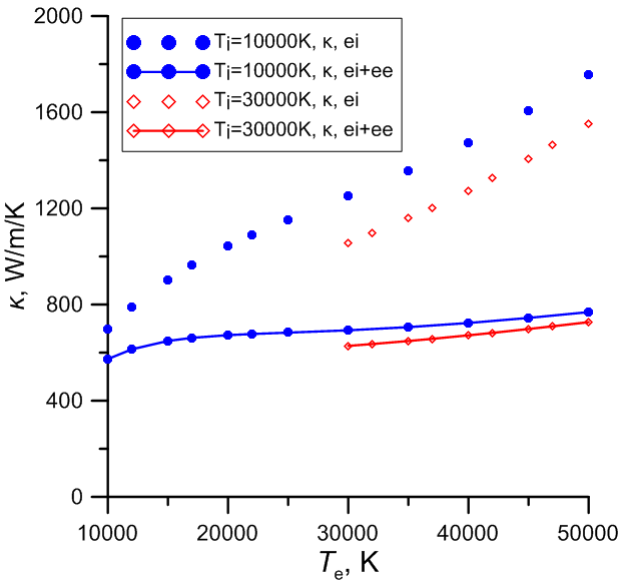


**Figure 6.** Electron thermal conductivity of liquid Al (at density of 2.35 g/cm$^3$) in dependence on the electron temperature at the temperatures of ions $T_\mathrm{i}$ = 10 000 and 30 000 K. Thomas–Fermi screening is used. Thermal conductivity coefficient is shown with only the electron–ion scattering taken into account and with the inclusion of electron–electron scattering.

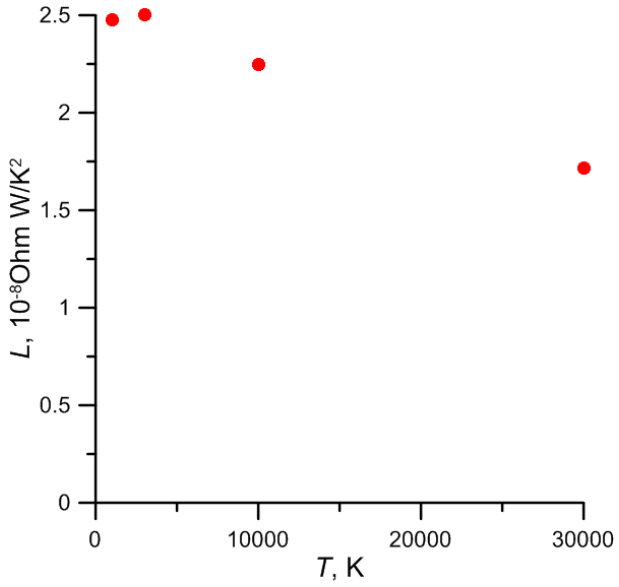


**Figure 7.** Lorenz number of liquid Al at the equilibrium one-temperature state in dependence on the temperature. Density is equal to 2.35 g/cm$^3$.

shown in figure 7. The two-temperature Lorenz number $L_\mathrm{e}(x, T_\mathrm{e}, T_\mathrm{i})$ as a function of electron temperature for several values of ion temperature is shown in figure 8. Both the equilibrium and the two-temperature Lorenz numbers were calculated with the use of Thomas–Fermi screening of the electron–ion interaction being taken into account.

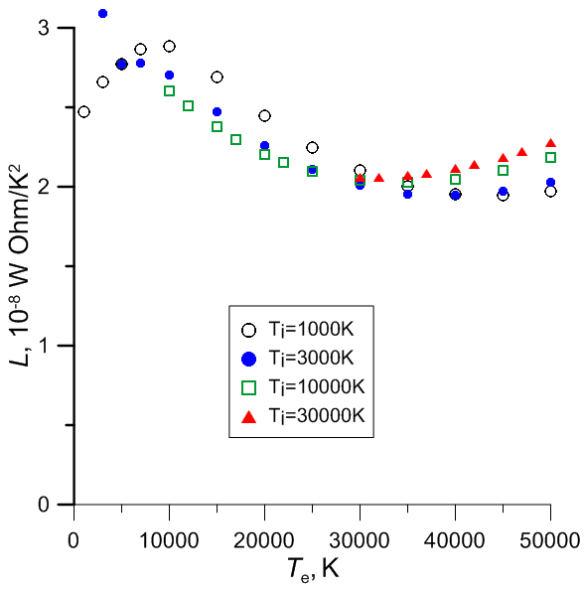


**Figure 8.** Two-temperature Lorenz number of the melt of aluminum depending on the electron temperature at the ion temperatures $T_i$ = 1000, 3000, 10 000, 30 000 K. Density of the melt is equal to 2.35 g/cm$^3$.

## 7. Conclusion

We have calculated electron kinetic coefficients of liquid aluminum in a framework of the Ziman approach. Main components of this approach are the structure factor of a liquid and the potential of the interaction between electrons and ions. We have calculated the structure factor by applying classical molecular dynamics with the previously obtained interatomic potentials. This method requires less computational effort with respect to the quantum molecular dynamics approach. When using electron–ion interaction different approximations for the electron screening of this interaction are considered. Resistivity and thermal conductivity of liquid aluminum are calculated both in the equilibrium states with equal temperatures of ions and electrons and in two-temperature states when the electron temperature exceeds the temperature of ions. It is shown that the thermal conductivity coefficient significantly decreases when the electron–electron scattering is taken into account. Lorenz number in the Wiedemann–Franz law both in the one-temperature and two-temperature states is obtained.

## Acknowledgments

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