Electrical resistivity and thermal conductivity of liquid aluminum in the two-temperature state

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Abstract. The electrical resistivity and thermal conductivity of liquid aluminum in the two-temperature state is calculated by using the relaxation time approach and structural factor obtained by molecular dynamics simulation.

1. Introduction

Electron heat conductivity is one of the most important intrinsic characteristics of the metal exposed to the ultrashort laser pulses. It significantly affects the rate of the heat propagation into the metal target and the thickness of the target heated layer. Resistivity and thermal conductivity at high temperatures are intensively investigated both by different aspects of Ziman or Kubo-Greenwood approach [1–5]. At the early stage of the laser action, when two-temperature state of a metal takes place, thermal conductivity depends on the electron and ion temperatures and density. Therefore kinetic coefficients of the two-temperature metals become important for the adequate description of the heat propagation in the laser target [6]. In [7–18] we have calculated thermal conductivity of some metals and their resistivity in the two-temperature state. In these works most attention was paid to the two-temperature state of solid metals while in the calculation of liquid state semiempirical approach was significantly used. In this work we apply the relaxation time approach with the use of structure factor obtained by using the molecular dynamics with many-body interaction between ions to get kinetic coefficients of aluminum in the two-temperature state.

2. Resistivity

The collision integral in the kinetic equation for the electrons can be written in standard form [19]:

$$\left.\frac{\partial f_{\mathbf{k}}}{\partial t}\right|_{\rm sc} = \int \left[f_{\mathbf{k}'}(1-f_{\mathbf{k}}) - f_{\mathbf{k}}(1-f_{\mathbf{k}'})\right] w(\mathbf{k},\mathbf{k}')\frac{V d\mathbf{k}'}{(2\pi)^3}$$

$$= \int \left(f_{\mathbf{k}'} - f_{\mathbf{k}}\right) w(\mathbf{k},\mathbf{k}')\frac{V d\mathbf{k}'}{(2\pi)^3},$$

where $f_{\mathbf{k}}, f_{\mathbf{k}'}$—non-equilibrium distribution function of electrons involved in a collision in the volume V of the metal, $w(\mathbf{k},\mathbf{k}')$—transition probability per unit time of the electron from the

state with wave vector $\mathbf{k}$ and the energy ε into the state with wave vector $\mathbf{k}'$ and the energy ε'. This probability in the Born approximation, taking into account the quasi-elastic scattering of the electron, can be written as

$$w(\mathbf{k},\mathbf{k}') = \frac{2\pi}{\hbar}\left|U_{\mathbf{k}\mathbf{k}'}\right|^2 \delta(\varepsilon-\varepsilon') = \tilde{w}(\mathbf{k},\mathbf{k}')\delta(\varepsilon-\varepsilon').$$

The matrix element of the interaction of electrons and ions of the metal $U_{\mathbf{k}\mathbf{k}'}$ assuming that the potential energy of the electron is the sum of the potential energies of interaction with individual ions with their concentration n_i, at the positions $\mathbf{R}_l$

$$U(\mathbf{r}) = \sum_l u(\mathbf{r}-\mathbf{R}_l),$$

can be represented as

$$U_{\mathbf{k}\mathbf{k}'} = \frac{n_i}{V} S(\mathbf{q},T_i,T_e,n_i)|u(\mathbf{q})|^2.$$

Here

$$u(\mathbf{q}) = u(q) = \int u(\mathbf{r}) e^{-i\mathbf{q}\mathbf{r}} d\mathbf{r}$$

is a Fourier transform of the pseudopotential $u(\mathbf{r})$ of the individual ion, $\mathbf{q} = \mathbf{k}' - \mathbf{k}$—change of momentum of the electron in the scattering.

$$S(\mathbf{q},T_i,T_e,n_i) = S(q,T_i,T_e,n_i) = \frac{1}{N}\overline{\sum_{\mathrm{ij}} e^{-i\mathbf{q}(\mathbf{R}_i-\mathbf{R}_j)}}$$

is a structural factor of the isotropic liquid, depending on the momentum transfer q, ion concentration, as well as on the ion temperature T_i and electron temperature T_e in non-equilibrium situation. Respectively

$$\tilde{w}(\mathbf{k},\mathbf{k}') = \tilde{w}(q) = \frac{2\pi}{\hbar}\frac{n_i}{V} S(q,T_i,T_e,n_i)|u(q)|^2$$

Within the relaxation time approximation, with the relaxation time depending on the energy of the particle, the collision integral is replaced by

$$\left.\frac{\partial f_{\mathbf{k}}}{\partial t}\right|_{\mathrm{sc}} = -\frac{f_{\mathbf{k}} - f_{\mathbf{k}0}}{\tau(\varepsilon)} = -\frac{f'_{\mathbf{k}}}{\tau(\varepsilon)}$$

with $f'_{\mathbf{k}} = f_{\mathbf{k}} - f_{\mathbf{k}0}$ and the equilibrium Fermi function $f_{\mathbf{k}0}$. At the relaxation time $\tau(\varepsilon)$ deviation from the equilibrium distribution function in the electric field $\mathbf{E}$ is $f'_{\mathbf{k}} = (-\partial f/\partial\varepsilon)\,\mathbf{v}_{\mathbf{k}} e\mathbf{E}\tau(\varepsilon)$. Substituting this expression into the collision integral in the form of

$$\left.\frac{\partial f_{\mathbf{k}}}{\partial t}\right|_{\mathrm{sc}} = \int \left(f'_{\mathbf{k}'} - f'_{\mathbf{k}}\right) w(q) \frac{V d\mathbf{k}'}{(2\pi)^3},$$

and equating the two forms of the collision integral representation within the approximation of an isotropic effective mass m with $\varepsilon(k) = \hbar^2 k^2/(2m)$ allows to find the electron relaxation time depending on the module k of its wave vector:

$$\tau(k) = \frac{2(2\pi)^2}{V} \cdot \frac{\hbar^2}{m} \frac{k^3}{\int_0^{2k} q^3 \tilde{w}(q) dq}.$$

Taking into account the expressions for $\tilde{w}(q)$ we get

$$\tau(k) = 4\pi \frac{\hbar^3}{m n_i} \frac{k^3}{\int_0^{2k} q^3 S(q, T_i, T_e, n_i) |u(q)|^2 dq}. \quad (1)$$

With the wave vectors k_F, taken only on the Fermi surface ($k_F = (3\pi^2 z n_i)^{1/3}$ with the number of conduction electrons per atom $z = 3$), in equilibrium ($T_e = T_i = T$) situation conductivity can be obtained as

$$\sigma(T, n_i) = \frac{n e^2 \tau(k_F)}{m} = 4\pi \frac{n e^2 \hbar^3}{m^2 n_i} \frac{k_F^3}{\int_0^{2k_F} q^3 S(q, T, n_i) |u(q)|^2 dq}.$$

Then the resistivity is

$$\rho(T, n_i) = \frac{1}{\sigma} = \frac{3\pi}{4} \frac{m^2 n_i}{e^2 \hbar^3 k_F^6} \int_0^{2k_F} q^3 S(q, T, n_i) |u(q)|^2 dq.$$

This is Ziman formula.

To account for the strong excitation of electrons above the Fermi energy ε_F and the appearence of a two-temperature situation at the interaction of femtosecond laser pulses with an aluminum target, we use the more general expression for the conductivity [19]:

$$\sigma = \frac{2}{3} e^2 \frac{4\pi}{(2\pi)^3} \int_0^\infty \tau(k) \left(-\frac{\partial f}{\partial \varepsilon} \right) \left(\frac{\hbar k}{m} \right)^2 k^2 dk. \quad (2)$$

Substituting here the relaxation time from (1), we get

$$\sigma(T_e, T_i, n_i) = \frac{4}{3\pi} \frac{\hbar^5 e^2}{m^3 n_i} \int_0^\infty \frac{k^7}{\int_0^{2k} q^3 S(q, T_i, T_e, n_i) |u(q)|^2 dq} \left(-\frac{\partial f}{\partial \varepsilon} \right) dk. \quad (3)$$

At low electron temperature $-\partial f/\partial \varepsilon = \delta(\varepsilon - \varepsilon_F) = m\delta(k - k_F)/(\hbar^2 k_F)$, and the expression for the conductivity (2) gives the Drude formula $\sigma = n e^2 \tau(k_F)/m$. In our two-temperature situation with the electron temperature T_e, higher than the temperature of the ions T_i, the expression for the $-\partial f/\partial \varepsilon$ is not reduced to δ-function and is

$$-\frac{\partial f}{\partial \varepsilon} = \frac{\exp\left(\dfrac{\varepsilon_0 + \hbar^2 k^2/(2m) - \mu}{k_B T_e} \right)}{\left(\exp\left(\dfrac{\varepsilon_0 + \hbar^2 k^2/(2m) - \mu}{k_B T_e} \right) + 1 \right)^2} \cdot \frac{1}{k_B T_e}$$

$$= \frac{1}{4 \cosh^2\left(\dfrac{\varepsilon_0 + \hbar^2 k^2/(2m) - \mu}{2 k_B T_e} \right)} \cdot \frac{1}{k_B T_e}.$$

Here $\varepsilon_0 = \varepsilon(n_i)$ is the energy of the conduction band (from our DFT-calculations at a density corresponding to zero temperature and zero pressure (2.75 g/cc), it is equal to $-\varepsilon_{F0} = -11.1$ eV, measured from the Fermi level taken as zero energy).

$\mu = \mu(T_e, n)$ is a chemical potential of the electrons. It is convenient to introduce the relative ion concentration relative to the concentration of ions n_{i0} at zero temperature and zero pressure:

$x = n_i/n_{i0}$. The value of n_{i0} corresponds to a density of 2.75 g/cc. Then at a constant effective mass of the electrons $\varepsilon_0(n_i) = -x^{2/3}\varepsilon_{F0}$, and $\mu(T_e, n)$ is the solution of equation

$$n = \frac{\sqrt{2}}{\pi^2}\left(\frac{\sqrt{m}}{\hbar}\right)^3 \int_0^\infty \frac{\sqrt{\varepsilon'}}{\exp\left(\dfrac{\varepsilon_0 + \varepsilon' - \mu}{k_B T_e}\right) + 1} d\varepsilon'.$$

We choose pseudopotential $u(r)$ necessary to calculate the electron relaxation time as a special case of Heine-Abarenkov pseudopotential with the screened Coulomb interaction

$$u(r) = -\xi\frac{ze^2}{r_0}\exp(-r_0/\lambda), r \le r_0$$

and

$$u(r) = -\frac{ze^2}{r}\exp(-r/\lambda), r > r_0.$$

and consider cases with $\xi = 0$ and 1. Here $\lambda = \lambda(T_e, x)$ is a screening length, r_0 is a core radius. The Fourier transform of the pseudopotential has a form

$$u(q) = 4\pi z e^2 r_0^2 \exp(-r_0/\lambda)\left(\xi\frac{\cos y - \sin y/y}{y^2} - \frac{\cos y + r_0 \sin y/(\lambda y)}{y^2 + (r_0/\lambda)^2}\right),$$

where $y = kr_0$. We calculate the inverse screening length of the Coulomb interactions in the Thomas-Fermi approximation as

$$\frac{1}{\lambda(T_e, x)} = \sqrt{\frac{4\pi e^2}{\partial\mu/\partial n_e}}.$$

Considering the asymptotic behavior of this expression at low and high temperatures, we use $\lambda(T_e, x)$ in the form

$$\lambda(T_e, x) = \sqrt{\frac{k_B T_e}{4\pi z n_i e^2}} + \sqrt{\frac{(-\varepsilon_0)x^{2/3}}{6\pi z n_i e^2}}.$$

Using the Fourier transform of the potential $u(q)$ and the calculated structure factor $S(q, T, T, n_i)$, the conductivity in the equilibrium case for the temperature T can be find from (3), and resistivity as the inverse of its value. The aluminum structure factor depends weakly on the electron temperature T_e because of the low dependence of interatomic interactions on the electron temperature [20–22]. Considering it depends only on the ion temperature and taking into account that the Fourier transform of the pseudopotential depends upon the electron temperature, we obtain in a two-temperature situation, when the electron temperature is different from the ion:

$$\sigma(T_e, T_i, n_i) = \frac{4}{3\pi}\frac{\hbar^5 e^2}{m^3 n_i}\int_0^\infty \frac{k^7}{\displaystyle\int_0^{2k} q^3 S(q, T_i, n_i)|u(q, T_e)|^2 dq}\left(-\frac{\partial f}{\partial\varepsilon}\right)dk. \tag{4}$$

The reverse of this value is the two-temperature resistivity:

$$\rho(T_e, T_i, n_i) = 1/\sigma(T_e, T_i, n_i) \tag{5}$$

Expression for the resistivity obtained by using (4) and (5) used by us differs from the often used Ziman-Evans expression [23, 24], which takes the form

$$\begin{aligned}\rho(T_e, T_i, n_i) &= \frac{4}{3\pi}\frac{m^2 e^2}{\hbar^3 n_i (4\pi z e^2)^2}\int_0^\infty \left(-\frac{\partial f}{\partial k}dk\right)\int_0^{2k} q^3 S(q, T_i, n_i)|u(q, T_e)|^2 dq \\ &= \frac{4}{3\pi}\frac{m^2 e^2}{\hbar^3 n_i (4\pi z e^2)^2}\int_0^\infty f\left(\frac{k}{2}\right)k^3 S(k, T_i, n_i)|u(k, T_e)|^2 dk \end{aligned} \tag{6}$$

in the two-temperature state.

3. Thermal conductivity

Electron relaxation time from (1), depending on its wave vector, can now be used to find the electron thermal conductivity due to the scattering of electrons by ions. In the absence of electric current, it can be found from the Onsager coefficients

$$I_0 = \frac{1}{3\pi^2\hbar^3 m_s^2}\int \frac{p^4}{\nu(p)}\left(-\frac{\partial f_s}{\partial \varepsilon}\right)dp,$$

$$I_1 = \frac{1}{3\pi^2\hbar^3 m_s^2}\int \frac{p^4}{\nu(p)}\left(-\frac{\partial f_s}{\partial \varepsilon}\right)(\varepsilon-\mu)dp,$$

$$I_2 = \frac{1}{3\pi^2\hbar^3 m_s^2}\int \frac{p^4}{\nu(p)}\left(-\frac{\partial f_s}{\partial \varepsilon}\right)(\varepsilon-\mu)^2 dp$$

as

$$\kappa_{\rm ei}(T_e, T_i, n_i) = \frac{1}{T_e}\left(I_2 - \frac{I_1^2}{I_0}\right).$$

At high electron temperatures the contribution of electron-electron collisions in the electronic conductivity significantly increases. Evaluating with the use of the Onsager coefficients the thermal conductivity due to electron-electron collisions $\kappa_{ee}(T_e, n_i)$ as in [18], we can now calculate the coefficient of electronic thermal conductivity as

$$\kappa(T_e, T_i, n_i) = \left(\kappa_{\rm ei}(T_e, T_i, n_i)^{-1} + \kappa_{\rm ee}(T_e, n_i)^{-1}\right)^{-1} \tag{7}$$

The calculated coefficient $\kappa_{\rm ee}(T_e, n_i)$ allows to calculatie the effective frequency of electron-electron collisions ν_{ee}, included in the Drude formula for thermal conductivity $\kappa_{\rm ee} = C_v v^2/\nu_{\rm ee}$ and depending on the electron temperature and density. Isochoric electron heat capacity in the Drude formula is calculated as

$$C_v = \frac{\sqrt{2}}{\pi^2}\left(\frac{\sqrt{m}}{\hbar}\right)^3 \int_0^\infty (\varepsilon_0+\varepsilon')\sqrt{\varepsilon'}\frac{1}{4\cosh^2\left(\dfrac{\varepsilon_0+\varepsilon'-\mu}{2k_BT_e}\right)}\frac{\mu_T T_e + \varepsilon_0 + \varepsilon' - \mu}{k_B T_e^2}d\varepsilon',$$

where

$$\mu_T = \frac{\partial \mu}{\partial T_e} = -\frac{1}{T_e}\frac{\displaystyle\int_0^\infty (\varepsilon_0+\varepsilon'-\mu)\sqrt{\varepsilon'}\cosh^{-2}\left(\frac{\varepsilon_0+\varepsilon'-\mu}{2k_BT_e}\right)d\varepsilon'}{\displaystyle\int_0^\infty \sqrt{\varepsilon'}\cosh^{-2}\left(\frac{\varepsilon_0+\varepsilon'-\mu}{2k_BT_e}\right)d\varepsilon'}$$

We introduce the variable $t = 6k_BT_e/(\varepsilon_{F0}x^{2/3})$ as in [11, 18]. Then the mean square velocity of the electrons can be represented as

$$v^2 = \frac{2\mu_0 x^{2/3}}{m}(1+t/4),$$

and the frequency of electron-electron collisions (in 10^{15} s^{-1}), derived from the results of calculation of $\kappa_{\rm ee}(T_e, n_i)$, can be approximated by the expression

$$\nu_{\rm ee}(T_e, x) = 1.40559x^{2/3}t^2\frac{1+0.307491t^{1.11513}}{1+0.45109t^{2.17729}}.$$

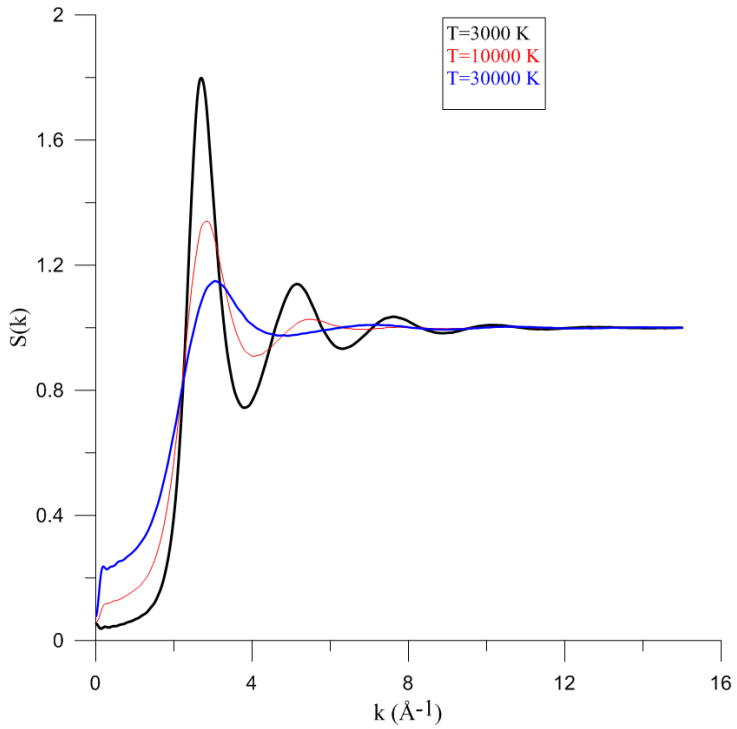

Figure 1. The structure factor of liquid aluminum in dependence of the wave number for three vallues of temperature

4. Results

The structure factor $S(k, T, n_i)$ in the equilibrium one-temperature situation is calculated within the molecular dynamics approach with the number of aluminum atoms equal to 13500 and 48688 (giving similar results) and the time step for the ion motion 1 fs. The many-body interatomic potential of the embedded atom model was taken as in [25,26]. Structure factor for several values of temperature T at the density 2.35 g/cc is shown in figure 1. As it was mentioned above, due to the weak dependence of the force constants of aluminum on the electron temperature within its range under consideration, the structure factor $S(k, T, n_i)$ can be used as the two-temperature structural factor $S(k, T_e, T_i, n_i)$ with $T_i = T$. The pseudopotential used has a free parameter r_0 which we choose to satisfy the well known value 0.24 μOhm·m of the resistivity of liquid aluminum at the melting temperature. We examined two limiting cases of the Heine-Abarenkov pseudopotential with $\xi = 0$ (it corresponds to the Ashkroft pseudopotential, and in this case $r_0 = 0.66$Å) and $\xi = 1$ (in this case $r_0 = 0.90$Å).

In figure 2 results of calculation of the resistivity of liquid aluminum in the equilibrium state with $T_e = T_i = T$ at the density 0.235 g/cc (which corresponds to the density in the liquid state in the melting) for the case ξ=1 are shown which are close to the data obtained in [4,27]. They are also very close to those obtained in [2]. Resistivity calculated by the use of the Ziman-Evans expression for it, is shown in figure 3 together with the used by us approach, when the resistivity is the reverse of the primarily calculated conductuvity. Both approaches give the same results at low temperatures but differ significantly with the electron temperature increase. Similar discrepancy between the reverse conductivity approach (4,5) and Ziman-Evans approach was found for the hydrogen plasma in [24].

Results obtained for the electron thermal conductivity of liquid aluminum in the equilibrium one-temperature state are shown in figure 4. Thermal conductivity increases with the electron

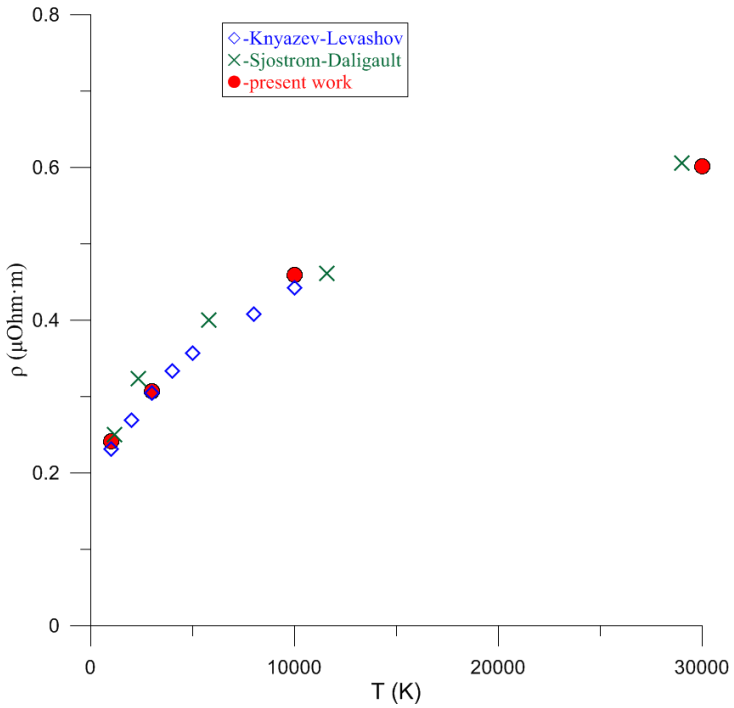

Figure 2. Resistivity of the liquid aluminum in the equilibrium state with the equal electron and ion temperatures $T_e = T_i = T$

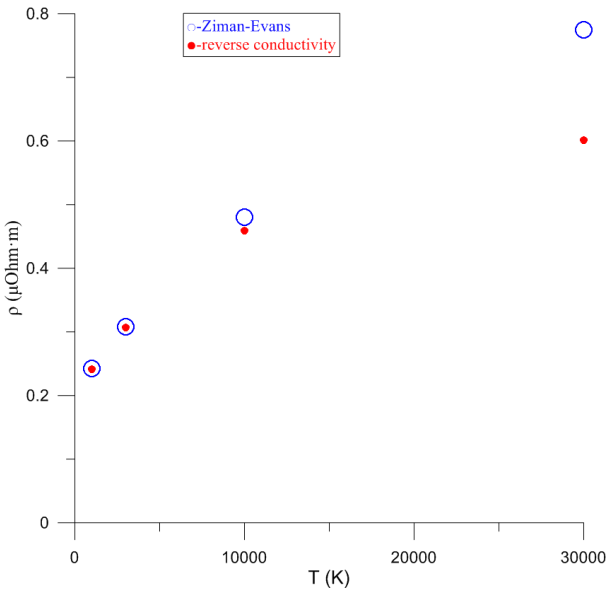

Figure 3. Comparison of the Ziman-Evans and reverse conductivity approach for the resistivity of the liquid aluminum in the equilibrium state with the common electron and ion temperatures $T_e = T_i = T$

temperature increase significantly slower when the electron-electron collisions are taken into account in addition to electron-ion collisions. Thermal conductivity of liquid aluminum in the two-temperature state is presented in figure 5. Due to the growth of the electron-electron collision

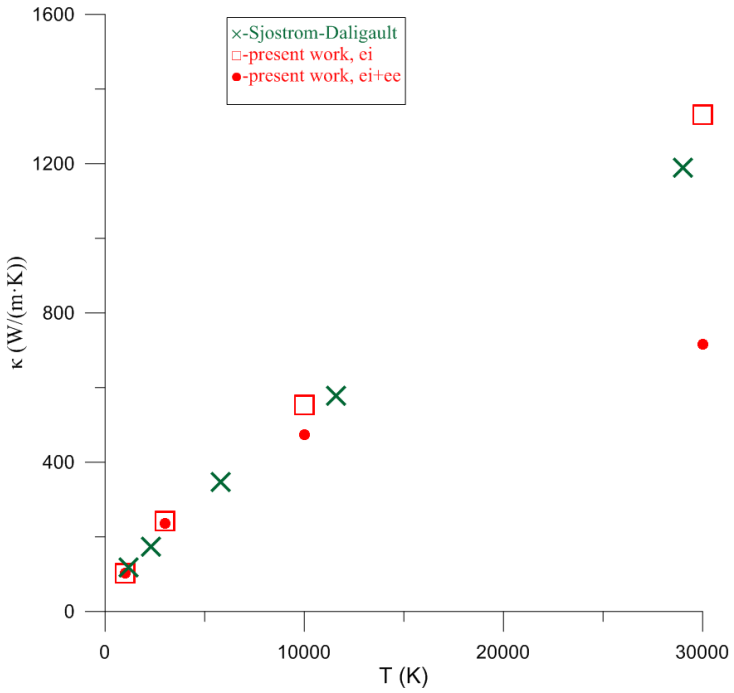

Figure 4. Thermal conductivity of the aluminum in the one-temperature liquid state in dependence on the temperature. Density is equal to 2.35 g/cc.

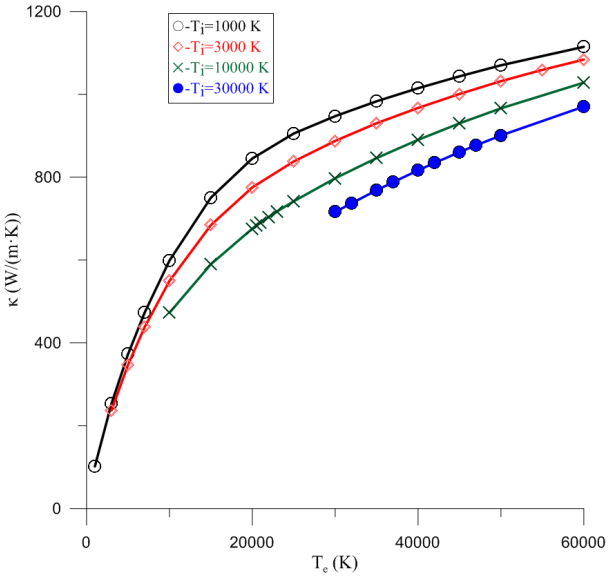

Figure 5. Two-temperature thermal conductivity of liquid aluminum at a density 2.35 g/cc. Dependence on the electron temperature T_e is shown for the values of ion temperature T_i=1000, 3000, 10000, 30000 K.

frequencies thermal conductivity is not growing strongly when the temperature of electrons increase. For the upper limit of electron temperature under consideration the excitation of L-electrons is negligibly small, and single electron energy band approach is applicable. Figure 6 presents the dependence of the resistivity on the electron temperature in the nonequilibrium

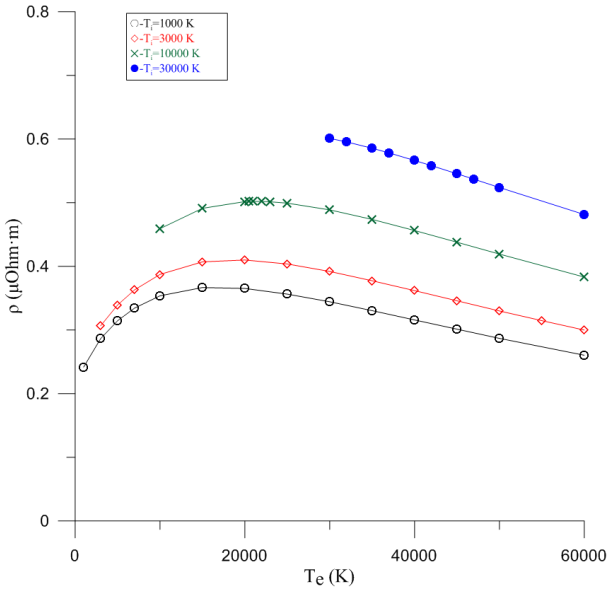

Figure 6. Resistivity of liquid aluminum in dependence of the electron temperature in the two-temperature states with the temperature of ions T_i=1000, 3000, 10000, 30000 K. Density in the liquid state is 2.35 g/cc.

two-temperature state of liquid aluminum for several values of the ion temperature.

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