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 of ions obtained by molecular dynamics simulation. Resistivity within the Ziman–Evans approach is also considered to be higher than in the approach with previously calculated conductivity via the relaxation time. Calculations based on the construction of the ion structural factor through the classical molecular dynamics and kinetic equation for electrons are more economical in terms of computing resources and give results close to the Kubo–Greenwood with the quantum molecular dynamics calculations.

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. This in turn determines the total pattern of ablation of a laser irradiated metal target [1–8]. Resistivity and thermal conductivity at high temperatures are intensively investigated both by different aspects of Ziman or Kubo–Greenwood approach [9–13]. 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 [14].

In [15–26], 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. Meanwhile, the intensities we are considering are such that the melting of the target material occurs [27, 28].

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. Aluminum belongs to the so-called simple metals and being widely used as a laser target, is intensively investigated both experimentally and theoretically [9, 10, 12, 29].
2. Resistivity
The collision integral in the kinetic equation for the electrons can be written in standard form [30]:

\[
\frac{\partial f_k}{\partial t} \bigg|_{sc} = \int [f_{k'}(1 - f_k) - f_k(1 - f_{k'})] w(k, k') \frac{V dk'}{(2\pi)^3}
\]

\[
= \int (f_{k'} - f_k) w(k, k') \frac{V dk'}{(2\pi)^3},
\]

where \(f_k\) and \(f_{k'}\) are the non-equilibrium distribution function of electrons involved in a collision in the volume \(V\) of the metal, \(w(k, k')\) is the transition probability per unit time of the electron from the state with wave vector \(k\) and the energy \(\varepsilon\) into the state with wave vector \(k'\) and the energy \(\varepsilon'\). This probability in the Born approximation, taking into account the quasi-elastic scattering of the electron when considering electron–ion interactions, can be written as

\[
w(k, k') = \frac{2\pi}{\hbar} |U_{kk'}|^2 \delta(\varepsilon - \varepsilon') = \tilde{w}(k, k')\delta(\varepsilon - \varepsilon').
\]

The matrix element of the interaction of electrons and ions of the metal \(U_{kk'}\) 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 \(R_i\)

\[
U'(r) = \sum_i u(r - R_i),
\]

can be represented as

\[
U_{kk'} = \frac{n_i}{V} S(q, T_i, T_e, n_i) |u(q)|^2.
\]

Here

\[
u(q) = u(q) = \int u(r)e^{-iqr}d\mathbf{r}
\]
is a Fourier transform of the pseudopotential \(u(r)\) of the individual ion, \(q = k' - k\) is the change of momentum of the electron in the scattering;

\[
S(q, T_i, T_e, n_i) = S(q, T_i, T_e, n_i) = \frac{1}{N} \sum_{ij} e^{-iq(R_i-R_j)}
\]
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 the non-equilibrium situation. Respectively

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

Within the relaxation time approximation, which is an important feature of Ziman approach, with the relaxation time depending on the energy of the particle, the collision integral is replaced by

\[
\frac{\partial f_k}{\partial t} \bigg|_{sc} = \frac{f_k - f_{k0}}{\tau(\varepsilon)} = -\frac{f'_k}{\tau(\varepsilon)}
\]

with \(f'_k = f_k - f_{k0}\) and the equilibrium Fermi function \(f_{k0}\). Calculations for the collisional plasma with arbitrary ionic charge based on the Fokker–Planck approach are made in [31]. At the relaxation time \(\tau(\varepsilon)\) deviation from the equilibrium distribution function in the electric field \(\mathbf{E}\) is \(f_k' = (-\partial f/\partial \varepsilon) \mathbf{v}_k e\mathbf{E}\tau(\varepsilon)\). Substituting this expression into the collision integral in the form of

\[
\frac{\partial f_k}{\partial t} \bigg|_{sc} = \int (f_{k'} - f_k') w(q) \frac{V dk'}{(2\pi)^3},
\]
and equating the two forms of the collision integral representation within the approximation of an isotropic effective mass $m$ with the electron energy $\varepsilon(k) = \hbar^2 k^2/(2m)$ allows to find the electron relaxation time depending on the module $k$ of its wave vector:

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

Energy of electron is measured from the bottom of the conduction band. Taking into account the electron relaxation time depending on the module $k$ of its wave vector:

$$\tau_{ei}(k) = 4\pi \frac{\hbar^3}{mn_i} \int_0^{2k} q^3 S(q, T, n_i)|u(q)|^2 dq.$$ 

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

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

Then the resistivity is

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

This is Ziman formula.

To account for the strong excitation of electrons above the Fermi energy $\varepsilon_F$ and the appearance 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 [30]:

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

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

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

At low electron temperature $-\partial f/\partial \varepsilon = \delta(\varepsilon - \varepsilon_F) = n\delta(k - k_F)/(\hbar^2 k_F)$, 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 $\delta$-function and is

$$-\frac{\partial f}{\partial \varepsilon} = \frac{\exp \left( \frac{h^2 k^2/(2m) - \mu}{k_B T_e} \right)}{\left( \exp \left( \frac{h^2 k^2/(2m) - \mu}{k_B T_e} \right) + 1 \right)^2 k_B T_e} = \frac{1}{4 \cosh^2 \left( \frac{h^2 k^2/(2m) - \mu}{2k_B T_e} \right) k_B T_e}.$$
\( \mu = \mu(T_e, n) \) is a chemical potential of the electrons. We suppose the effective mass of the electrons to be constant. It can be obtained from the low temperature and zero pressure DFT-calculations of the electron spectra and density of states [32]. Fermi level at these conditions corresponds to \( \varepsilon_{F0} = 11.1 \) eV and effective mass can be obtained as

\[
m = \frac{\hbar^2 (3\pi^2 z n_{\text{i}0})^{2/3}}{2\varepsilon_{F0}}
\]

with the ion concentration \( n_{\text{i}0} = 2.75 \) g/cm\(^3\). Thus calculated effective mass is 1.06 of the free electron mass. Then the chemical potential \( \mu(T_e, n) \) is the solution of equation

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

It is convenient to introduce relative ion concentration: \( x = n_1/n_{\text{i}0} \). We choose pseudopotential \( u(r) \) necessary to calculate the electron relaxation time as a special case of Ashcroft pseudopotential with the screened Coulomb interaction

\[
u(r) = \begin{cases} 0, & r \leq r_0 \\ \frac{z^2 e^2}{r} \exp(-r/\lambda), & r > r_0 \end{cases}
\]

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

\[
u(q) = -4\pi z e^2 r_0^2 \cos(y) + r_0 \sin(y/(\lambda y))
\]

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

\[
\lambda(T_e, x) = \sqrt{\frac{k_B T_e}{4\pi z n_1 e^2}} + \sqrt{\frac{\varepsilon_{F0} x^{2/3}}{6\pi z n_1 e^2}}.
\]

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 [33–35]. 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:

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

with Fourier transform of potential given by (4). The reverse of value (5) is the two-temperature resistivity:

\[
\rho(T_e, T_i, n_i) = 1/\sigma(T_e, T_i, n_i).
\]
The calculated coefficient 

\[ \kappa_{ee}(T_e, n_i) = (\kappa_{ei}(T_e, T_i, n_i))^{-1} \]

and depending on the electron temperature and density. Isochoric electron heat capacity in the Drude formula is calculated as

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

where

\[ \mu_T = \frac{\partial \mu}{\partial T_e} = - \frac{1}{T_e} \int_0^{\infty} (\varepsilon - \mu) \sqrt{\varepsilon} \cosh^{-2} \left( \frac{\varepsilon - \mu}{2k_B T_e} \right) \, d\varepsilon. \]

We introduce the variable \( t = 6k_B T_e/(\varepsilon_{F0} x^{2/3}) \) as in [19, 26]. Then the squared velocity of electrons in Drude formula as Fermi velocity squared at low temperatures and Maxwell velocity squared at the high temperature limit can be represented as

\[ v^2 = \frac{2\mu_0 T_e x^{2/3}}{m} (1 + t/4), \]
Figure 1. The structure factor of liquid aluminum in dependence of the wave number for three values of temperature.

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

$$\nu_{ee}(T_e, x) = 1.40559x^{2/3}t^{1.1513} + 0.307491t^{1.11513} + 0.45109t^{2.17729}.$$  

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 [39, 40]. Structure factor for several values of temperature $T$ at the density 2.35 g/cm$^3$ which corresponds to the density in the liquid state in the melting 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 as $r_0 = 1.08$ a.u. (1 a.u. = 0.529 Å is the Bohr radius) to satisfy the well known value 0.24 $\mu$Ohm m of the resistivity of liquid aluminum at the melting temperature.

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 2.35 g/cm$^3$ are shown which are close to the data obtained in [12, 41]. They are also very close to those obtained in [10].
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 $T_e = T_i = T$. 
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/cm$^3$.

Figure 5. Two-temperature thermal conductivity of liquid aluminum at a density 2.35 g/cm$^3$. Dependence on the electron temperature $T_e$ is shown for the values of ion temperature $T_i = 1000, 3000, 10,000$ and $30,000$ K.
Figure 6. Resistivity of liquid aluminum in dependence on the electron temperature in the two-temperature states with the temperature of ions $T_i = 1000, 3000, 10000$ and $30000$ K. Density in the liquid state is $2.35$ g/cm$^3$.

Resistivity calculated by the use of the Ziman–Evans expression, is shown in figure 3 together with the approach used here, when the resistivity is the reverse of the primarily calculated conductivity. Both approaches give the same results at low temperatures but differ significantly with the electron temperature increase. Similar discrepancy between the reverse conductivity approach based on the equations (5) and (6) and Ziman–Evans approach was found for the hydrogen plasma in [37].

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

5. Conclusion
We have considered kinetic coefficients of liquid aluminum by using the $\tau$-approach of the kinetic equation. With the use of Ziman approximation applying the structure factor to relaxation time of electron (depending on the wavevector) the conductivity is calculated. Resistivity is then obtained as the reverse conductivity. Calculating the Onsager coefficients within the $\tau$ approach, the thermal conductivity has been found. Resistivity and thermal conductivity in the equilibrium one-temperature states agree well with the results of calculations based on the quantum molecular dynamics and the Kubo–Greenwood formalism. Resistivity and thermal
conductivity in two-temperature states, which arise at the interaction of femtosecond laser pulses with metals, are calculated at temperature of electrons higher than that of ions.

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References
[1] Anisimov S I, Zhakhovskii V V, Inogamov N A, Nishihara K, Petrov Yu V and Khokhlov V A 2006 J. Exp. Theor. Phys. 103 183–97
[2] Inogamov N A et al 2013 Contrib. Plasma Phys. 53 796–810
[3] Agranat M B et al 2007 JETP Lett. 85 271–6
[4] Povarnitsyn M E, Itina T E, Khishchenko K V and Levashov P R 2007 Appl. Surf. Sci. 253 6343–6
[5] Povarnitsyn M E, Itina T E, Sentis M, Khishchenko K V and Levashov P R 2007 Phys. Rev. B 75 235414
[6] Veysman M E, Agranat M B, Andreev N E, Ashitkov S I, Fortov V E, Khishchenko K V, Kostenko O F, Levashov P R, Ovchinnikov A V and Sitnikov D S 2008 J. Phys. B 41 125704
[7] Povarnitsyn M E, Itina T E, Levashov P R and Khishchenko K V 2011 Appl. Surf. Sci. 257 5168–71
[8] Andreev N E et al 2015 Laser Part. Beams 33 541–50
[9] Desjarlais M P, Kress J D and Collins L A 2002 Phys. Rev. E 66 025401
[10] Recoules V and Crocombette J P 2005 Phys. Rev. B 72 104202
[11] Faussurier G, Blancard C, Combis P and Videau L 2014 Phys. Plasmas. 21 092706
[12] Sjostrom T and Daligault J 2015 Phys. Rev. E 92 063304
[13] Ovechkin A A, Loboda P A and Falkov A L 2016 High Energy Density Phys. 20 38–54
[14] Ng A, Sterne P, Hansen S, Recoules C, Chen Z, Tsui Y Y and Wilson B 2016 Phys. Rev. E 94 032213
[15] Inogamov N A and Petrov Yu V 2010 J. Exp. Theor. Phys. 110 446–68
[16] Inogamov N A et al 2012 AIP. Conf. Proc. 1464 593–608
[17] Migdal K P, Petrov Y V and Inogamov N A 2013 Proc. SPIE 9065 906503
[18] Petrov Yu V and Inogamov N A 2013 JETP Lett. 98 278–84
[19] Petrov Yu V, Inogamov N A and Migdal K P 2013 JETP Lett. 97 20–7
[20] Petrov Yu V, Inogamov N A, Anisimov S I, Migdal K P, Khokhlov V A and Khishchenko K V 2015 J. Phys.: Conf. Ser. 653 012087
[21] Migdal K P, Il‘nitsky D K, Petrov Yu V and Inogamov N A 2015 J. Phys.: Conf. Ser. 653 012086
[22] Petrov Yu V, Inogamov N A and Migdal K P 2015 PIERS Proceedings 2431–5
[23] Petrov Yu V, Migdal K P, Knyazev D V, Inogamov N A and Levashov P R 2016 J. Phys.: Conf. Ser. 774 012013
[24] Petrov Y V, Khokhlov V A, Inogamov N A, Khishchenko K V and Anisimov S I 2016 J. Phys.: Conf. Ser. 774 012099
[25] Migdal K P, Petrov Y V, Il‘nitsky D K, Zhakhovsky V V, Inogamov N A, Khishchenko K V, Knyazev D V and Levashov P R 2016 Appl. Phys. A 122 408
[26] Petrov Y V, Migdal K P, Inogamov N A and Anisimov S I 2016 JETP Lett. 104 431–9
[27] Inogamov N A, Zhakhovskii V V, Ashitkov S I, Khokhlov V A, Petrov Yu V, Komarov P S, Agranat M B, Anisimov S I and Nishihara K 2009 Appl. Surf. Sci. 255 9712–6
[28] Povarnitsyn M E, Khishchenko K V and Levashov P R 2009 Appl. Surf. Sci. 255 5120–4
[29] Komarov P S, Ashitkov S I, Ovchinnikov A V, Sitnikov D S, Veysman M E, Levashov P R, Povarnitsyn M E, Agranat M B, Andreev N E, Khishchenko K V and Fortov V E 2009 J. Phys. A 42 214057
[30] Abrikosov A A 1988 Fundamentals of the Theory of Metals (Amsterdam: North-Holland)
[31] Nersisyan H B, Veysman M E, Andreev N E and Matevosyan H H 2014 Phys. Rev. E 89 033102
[32] Petrov Yu V, Migdal K P, Inogamov N A and Zhakhovsky V V 2015 Appl. Phys. B 119 401–11
[33] Medvedev D M and Petrov Yu V 1999 J. Exp. Theor. Phys. 88 128–34
[34] Minakov D V and Levashov P R 2015 Phys. Rev. B 92 224102
[35] Harbour L, Dharma-wardana M W C, Klug D D and Lewis L J 2015 Contrib. Plasma Phys. 55 144–51
[36] Starrett C E 2016 High Energy Density Phys. 19 58–64
[37] Burritt D J, Feinblum D V, Charest M R J and Starrett C E 2016 High Energy Density Phys. 19 1–10
[38] Ashcroft N W and Mermin N D 1976 Solid State Physics (New York: Holt, Rinehart and Winston)
[39] Winey J M, Kubota A and Gupta Y M 2009 Modell. Simul. Mater. Sci. Eng. 17 055004
[40] Winey J M et al 2010 Modell. Simul. Mater. Sci. Eng. 18 029801
[41] Knyazev D V and Levashov P R 2014 Phys. Plasmas 21 073302