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Electrical conductivity of dense aluminium fluid

S Kuhlbrodt and R Redmer

Department of Physics, University of Rostock, 18051 Rostock, Germany

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Abstract

Experiments using the rapid wire evaporation technique provided data for the electrical conductivity of aluminium plasmas in a density range of (0.001–2.3) g cm^{-3} and for temperatures between 10 000 and 285 000 K.

Within the frame of the linear response theory in the formulation of Zubarev, calulations of the electrical conductivity were made in the same density and temperature area. The composition of the plasma was determined within the partially ionized plasma model; the interaction between particles is considered to be a Debye potential (charged particles) and a screened polarization potential (interaction with atoms). The agreement between experimental and theoretical data is reasonable for arbitrary temperatures and densities $\varrho < 0.7$ g cm^{-3}.

For higher densities, in the warm dense fluid region, strong correlation effects occur. These effects can be taken into account including local field corrections in the dielectric function, using a dynamic ion–ion structure factor, or by replacing the Debye potential of the electron–ion interaction with a weak pseudopotential. The latter can be determined within a density functional theory. The influence of these effects on the electrical conductivity of a warm dense aluminium fluid is shown.

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1. Introduction

Transport properties of a plasma are strongly correlated to its state. Electrical conductivity as one of these properties plays an important role in determining plasma density and temperature.

Using the rapid wire evaporation technique, experiments provided data for the electrical conductivity for various metal plasmas, see [1–5]. In these experiments, the metals undergo a transition from a warm dense fluid with strong coupling and a degenerate electron system to a weakly coupled plasma where electrons can be treated classically.

Standard methods such as the Spitzer theory [6] or the original Ziman theory [7] cannot describe both cases. The challenge for the theory of plasmas is to reproduce the experimental data in the whole studied density–temperature region, which is largest for aluminium: $0.001\ \mathrm{g\ cm^{-3}} < \varrho < 2.3\ \mathrm{g\ cm^{-3}}$ and $8000\ \mathrm{K} \leqslant T \leqslant 285\,000\ \mathrm{K}$.

2. Electrical conductivity within LRT

The linear response theory (LRT) in the formulation of Zubarev [8] is a general approach to transport properties of plasmas on the basis of the correlation function method. It is valid for arbitrary degeneracy; the Spitzer results and the Ziman theory can be reproduced as limiting cases [9, 10].

Within this method, the electrical conductivity σ is given in a determinant representation

$$\sigma = \frac{-e^2}{\Omega_0 \left|(D_{nm})\right|} \begin{vmatrix} 0 & (Q_{0m}) \\ (N_{n0}) & (D_{nm}) \end{vmatrix} \tag{1}$$

where Ω_0 is the system volume, N_{n0}, Q_{0m} and D_{nm} $(n, m = 0, 1, 2, \ldots)$ are correlation functions of generalized momenta of the electron system.

N_{nm} and Q_{nm} depend on the number of free electrons. D_{nm} can be separated with respect to electron–electron, electron–ion, and, if atoms occur, electron–atom contributions $D_{nm} = D^{\mathrm{ee}}_{nm} + D^{\mathrm{ei}}_{nm} + D^{\mathrm{ea}}_{nm}$ which are, in turn, related to the transport cross sections Q^{ec}_T, $c = \{\mathrm{e}, \mathrm{i}, \mathrm{a}\}$. Depending on the strength of scattering, Q^{ec}_T can be evaluated in the T matrix or in the Born approximation with respect to the relevant interaction.

For more details see [10–12].

3. Results for the plasma region

For low densities, the *partially ionized plasma model* (PIP) based on the chemical picture with well-defined particles is an appropriate model. Bound states such as atoms are treated on the same level as the elementary particles electrons and protons.

For an aluminium plasma we consider free electrons e, different species of ions Al^{k+} up to a charge of $k = 5$, and neutral atoms Al^0. These particles are connected via chemical reactions in equilibrium, which leads to a system of coupled mass action laws. Together with the neutrality condition $n_e = \sum_{k=1}^{5} k n_k$, this system can be solved to derive the partial densities n_k of each species considered, see [12].

The electrical conductivity, thermal conductivity and thermopower of various metals were calculated for such a PIP plasma [12]; figure 1 shows the results for the electrical conductivity σ of aluminium.

Since strong scattering might occur between charges, Q^{ei}_T and Q^{ee}_T were evaluated on T matrix level with respect to a Debye potential V_{D}. This reproduces the Spitzer values in the low-density limit. The screened polarization potential between electrons and atoms is weak, so the Born approximation is sufficient for Q^{ea}_T. The minimum behaviour of the electrical conductivity for increasing densities and low temperatures of about 10 000 K is due to relocalization of free electrons and pressure ionization afterwards.

The overall agreement with the experimental data is good for densities $\varrho \leqslant 0.7\ \mathrm{g\ cm^{-3}}$ and arbitrary temperatures. Especially, the sharp increase in σ for $\varrho \geqslant 0.1\ \mathrm{g\ cm^{-3}}$ at 10 000 K can be reproduced. The deviations between the 10 000 K-isotherm and Krisch and Kunze data at this temperature can be explained with uncertainties in the temperature measurement.

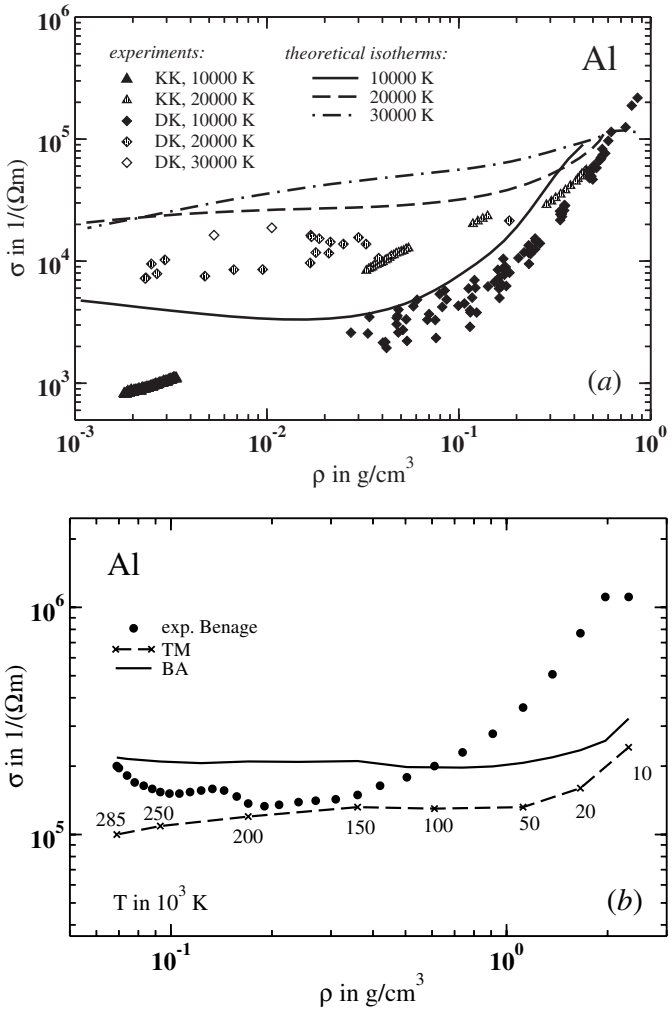

Figure 1. LRT results for the plasma region compared with experimental data. (*a*) shows isotherms for 10 000 K, 20 000 K and 30 000 K, and experimental results of Krisch and Kunze (KK) [1], and DeSilva and Katsouros (DK) [2]. (*b*) shows measured data by Benage *et al* [3]; theoretical curves are calculated at the same densities and temperatures as in that experiment. Transport cross sections are always calculated in T matrix approximation (TM). In addition, the results for the Born approximation (BA) are given in (*b*).

In the high-density limit, σ is independent of temperature. Therefore, the differences between theoretical and experimental data should be small at the highest densities.

4. The high-density case

The deviations in figure 1(*b*) at $\varrho \geqslant 1.0$ g cm^{-3} are a result of the model used so far. In these conditions, aluminium plasma is in the warm, dense matter regime. A physical picture with ions immersed in a degenerate electron gas is more reliable there (see, e.g., [13]). Furthermore, correlations in the ion system and in the electron gas occur, and the simple Debye potential is not a good approximation there.

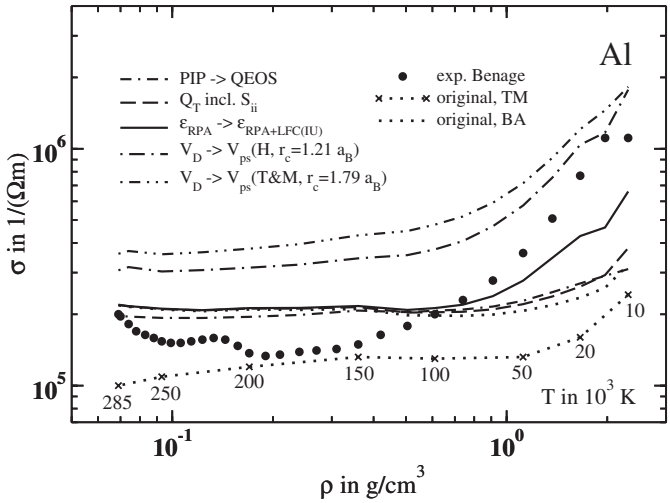

Figure 2. Influence of different improvements made to the 'original' calculation (see figure 1). (— · — — · —): PIP composition was replaced by QEOS composition. (— — —): a hard sphere structure factor was included. (——): LFC by Ichimaru–Utsumi [18] was included in screening. (— · — and — · · —): the Debye potential was replaced by a pseudopotential (H: Hamann scheme, T&M: Troullier–Martins scheme). For comparison, experimental data of Benage *et al* [3] (•), and the 'original' curves from figure 1(*b*) are included (dotted lines).

Since screening is strong at high densities, the Born approximation is suitable for the calculation of transport cross sections

$$Q_T^{\mathrm{ei}}(k) = \frac{4\pi}{k^4} \int_0^{2k} \mathrm{d}q\, q^3 \left| \frac{V_{\mathrm{ei}}(q)}{\varepsilon(q)} \right|^2 S_{\mathrm{ii}}(q). \tag{2}$$

Electron–electron scattering is of minor importance due to Pauli blocking, and atoms vanish at high densities due to pressure ionization. This makes it easy to take into account the correlations mentioned above: as a first step, ion–ion correlations are included in the static structure factor S_{ii}, electron–electron correlations can be considered in the screening of the potential $V_{\mathrm{ei}}(q)$ via local field corrections, and even the possibility of using a pseudopotential, as known from condensed matter physics, is given.

The input of another composition is also no problem, even if it is derived within a physical picture: ions can be treated as particles with an average rational charge that represents the mean ionization degree per heavy particle.

Figure 2 shows the influence of these possible improvements of the theory. As in figure 1(*b*), all curves are calculated at the same densities and temperatures as given by the experiment. Transport cross sections were calculated in the Born approximation (2) (BA). Electron–electron scattering is still included on the BA level, but has no influence at the highest densities.

The QEOS model [14] and its numerical conversion MPQeos [15], which was used as an example for a different equation of state and composition [16], are based on the Thomas–Fermi model, but include semi-empirical bonding corrections, and the ion thermal motion determined from solid or liquid state models. In contrast to PIP, QEOS was developed to describe warm dense matter. But that composition also cannot reproduce the experimental data (see figure 2). Even if one considers a plasma with only Al^{3+} ions for the highest density $\varrho = 2.3$ g cm^{-3} and 10 000 K, as found in molecular dynamic simulations, the electrical conductivity just reaches

values of $\sigma^{\mathrm{TM}} = 1.87 \times 10^5\ (\Omega\,\mathrm{m})^{-1}$ and $\sigma^{\mathrm{BA}} = 2.84 \times 10^5\ (\Omega\,\mathrm{m})^{-1}$, respectively, which is much lower than the electrical conductivity measured in the experiment [3]. Therefore, it can be stated that the influence of the composition on the electrical conductivity is small at high densities.

The effect of ion–ion correlations is also small. As a first attempt, the hard sphere structure factor $S_{\mathrm{ii}}^{\mathrm{HS}}$ with a radius $r_{\mathrm{HS}} = 0.054$ nm (the radius of Al^{3+} ions in a crystal [17]) was used. Including $S_{\mathrm{ii}}^{\mathrm{HS}}$, σ reaches higher values, but the increase with the density is still not strong enough compared with the experimental data.

The largest effect on electrical conductivity is the replacement of the Debye potential V_{D} by an effective electron–ion interaction. For this, electron–electron correlations can be included in the screening function considering local field corrections (LFC). The Debye potential used so far coincides with the static screening function $\varepsilon = \varepsilon_{\mathrm{RPA}}(q)$ in (2), which is now replaced by $\varepsilon_{\mathrm{RPA+LFC}}(q)$. We used the approved LFC by Ichimaru and Utsumi [18].

Another possibility of using an effective electron–ion potential is replacing V_{D} by a weak pseudopotential. For instance, the code fhi98PP [19] provides constructions of pseudopotentials within the Hamann scheme [20] and the Troullier–Martins scheme [21] with different approximations for the exchange and correlation functionals. For the results shown in figure 2, the local density approximation (LDA) by Perdew and Wang [22] was used, the cut-off radii r_{c} are given by default. Using generalized gradient approximation (GGA) [23] or exact exchange (EXX) [24] gives similar results as when using LDA.

For both ways it is found that σ increases up to values similar to the experimental ones in the high-density region (see figure 2). Using LFC has the advantage that it is valid over the whole density region shown here. For the pseudopotentials, a crossover to the Debye potential at low densities is needed.

5. Summary

Within the simple plasma approach as described in sections 2 and 3, the calculated results of the electrical conductivity of aluminium plasma are in good agreement with experimental data for densities $\varrho \leqslant 0.7$ g cm^{-3} and the whole temperature region of 10 000 K$\leqslant T \leqslant$ 285 000 K. For higher densities, the inclusion of high-density effects is possible. The effect of a different composition on electrical conductivity is small. The same can be said about the influence of an ion–ion structure factor. The largest effect is the change of the potential from Debye to an effective interaction. LFC can be used over the whole density region. When using a pseudopotential, a crossover to the Debye potential is needed for low densities. Self-consistent approaches to these effects (composition, structure factor and effective electron–ion interaction) are successful in calculating the electrical conductivity of plasmas, see [13, 25, 26]. A self-consistent use of high-density effects in LRT, including better approximations for S_{ii}, will provide reliable data not only for the electrical conductivity in a large density–temperature region, but also for thermal conductivity and thermopower.

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