Calculation of ionization balance and electrical conductivity in nonideal aluminum plasma

Deok-Kyu Kim* and Inho Kim
Agency for Defense Development, P.O. Box 35-5, Yuseong, Daejeon 305-600, Korea
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A practical approach has been implemented to calculate the ionization balance and electrical conductivity of warm dense aluminum plasma with the Coulomb coupling effect taken into account. The correction term for ionization potential is formulated with a number of basic dimensionless parameters that characterize nonideal plasma and incorporated with the fitted formulas of excess free energy given by Tanaka, Mitake, and Ichimaru [Phys. Rev. A **32**, 1896 (1985)] and Chabrier and Potekhin [Phys. Rev. E **58**, 4941 (1998)] to determine the ionization balance in an equilibrium state. The calculated degree of ionization of aluminum plasma exhibits a sudden increase near the solid density $\sim 1\ \mathrm{g/cm^3}$ at temperatures of a few eV, which effectively demonstrates the pressure-induced ionization. The electrical conductivity is evaluated in a partially ionized plasma regime based on a linear mixture rule that takes into account both the electron-ion and electron-neutral collisions and then the computed results are compared with available data from recent experiments. It is shown that the calculation well reproduces the overall trend of measured electrical conductivity of nonideal aluminum plasma accounting for the metal-insulator transition.

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I. INTRODUCTION

Nonideal plasma is characterized by strong Coulomb couplings between charged particles, which significantly affect the ionization balances, and frequently encountered in matters under extreme conditions such as the interior materials of a star or a heavy planet [1], shock-induced plasmas [2,3], and exploding metal wires [4]. A key parameter that measures the degree of nonideality of plasma is the Coulomb coupling constant which is defined as the ratio of the average Coulomb energy between charged particles to the average thermal energy. In the case where the Coulomb coupling constant remains small enough, the classical Debye-Hückel theory can be applied to describe the plasma condition [5]. However, when the Coulomb interaction energy becomes comparable to the thermal energy, the classical theory is no longer valid and thus the strong Coulomb coupling effect should be introduced in the theory. To date, many efforts have been made to provide comprehensive knowledge and physical insight of nonideal plasmas on the theoretical basis [6–12].

In recent years, the electrical conductivities of nonideal metal plasmas have been successfully measured by many authors in the regime of metal-insulator transition owing to the advanced experimental techniques such as the short-pulse laser heating and the capillary wire discharge of solid metals [13–18], which also enabled implementation of a semiempirical pressure-ionization model for practical applications [19]. However, there are only a limited number of theoretical models available for comparisons with the measured data in a wide density-temperature domain since most theoretical studies that involve self-consistent approaches mainly based on the quantum statistical theory or standard kinetic theory are subject to restrictions on their valid parameter space accompanied by inherent complexities [20–25]. It is essentially required to have a wide-range model with a full consideration of the Coulomb coupling effect on the ionization balance for extensively exploring the nonideal plasma properties.

In this paper, we describe an approach practically useful to calculate the electrical conductivity of nonideal plasma over a large density-temperature space by introducing a generalized formulation of the corrected potential term for ionization balances in an equilibrium state and then present comparisons of the calculated results with recent experimental data for aluminum. In formulating the corrected potential term, we make use of the fitted formulas of excess free energy given as functions of basic dimensionless parameters by Tanaka, Mitake, and Ichimaru [9] and Chabrier and Potekhin [12], which have been derived from massive analytical and computational procedures including Monte Carlo simulations and hypernetted-chain (HNC) calculations. Evaluation of the electrical conductivity in partially ionized aluminum plasma is based on a linear mixture rule in the form of additive collision frequencies for fully and weakly ionized plasmas, which proves efficient with the reasonably acceptable accuracy and minimized computational effort. The validity of the physical model employed in this work is to be verified through the comparisons between calculations and measurements.

II. IONIZATION BALANCE

The condition of nonideal plasma can be described with a number of dimensionless parameters. The Coulomb coupling constant is generally used to measure the correlated strength between charged particles in plasma and is defined for both ion and electron, respectively, by

$$\Gamma_i = \frac{Z_{eff}^2 e^2}{4\pi\epsilon_0 a_i}\frac{1}{k_B T} = \frac{(\text{interionic Coulomb energy})}{(\text{thermal energy})}, \quad (1a)$$

*Electronic address: dkyukim@add.re.kr

$$\Gamma_e = \frac{e^2}{4\pi\epsilon_0 a_e}\frac{1}{k_B T} = \frac{(\text{interelectronic Coulomb energy})}{(\text{thermal energy})}, \tag{1b}$$

where Z_{eff} is the effective charge number of plasma ions, $a_i = (\frac{4}{3}\pi n_i)^{-1/3}$, and $a_e = (\frac{4}{3}\pi n_e)^{-1/3}$ are the average distances between ions and electrons, respectively, known as the Wigner-Seitz radii, n_i is the total number density of ions, and n_e is the electron number density. Also useful is the electron density parameter

$$r_s = a_e / a_B \tag{2}$$

to represent the density effect, where $a_B = 4\pi\epsilon_0 \hbar^2 / m_e e^2$ is the Bohr radius and m_e is the electron mass. In addition, since the electron degeneracy becomes important in dense plasma, it is necessary to introduce the Fermi degeneracy parameter

$$\theta = k_B T / E_F, \tag{3}$$

where $E_F = \hbar^2 (3\pi^2 n_e)^{2/3} / 2m_e$ is the Fermi energy of electron.

These dimensionless parameters are commonly used to give generalized expressions of the excess free energy or the internal energy for nonideal plasmas with the mathematical complexity significantly reduced [9–12]. The total excess free energy can be decomposed into three independent terms each associated with different type of Coulomb correlation: ion-ion (i-i), ion-electron (i-e), and electron-electron (e-e) interactions. Here, the excess part of Helmholtz free energy is then written as

$$F_C = F_C^{i\text{-}i} + F_C^{i\text{-}e} + F_C^{e\text{-}e} = N_i k_B T [f_C^{i\text{-}i}(\Gamma_i) + f_C^{i\text{-}e}(\Gamma_e, r_s)] + N_e k_B T f_C^{e\text{-}e}(\Gamma_e, \theta), \tag{4}$$

where N_i and N_e are the total number of ions and electrons, respectively, $f_C^{i\text{-}i}$, $f_C^{i\text{-}e}$, and $f_C^{e\text{-}e}$ are the excess free energy density functions of corresponding interaction types which are reduced to dimensionless expressions.

The equilibrium ionization balance equation of an atomic element M for its interaction $M^k \leftrightarrow M^{k+1} + e^-$ is derived from a mass action law following the minimum free energy principle to be

$$\frac{n_{k+1} n_e}{n_k} = 2\left(\frac{m_e k_B T}{2\pi\hbar^2}\right)^{3/2} \frac{U_p^{k+1}}{U_p^k} \exp\left(-\frac{I_{k+1} - \Delta I_{k+1}}{k_B T}\right), \tag{5}$$

where $k = 0, \ldots, Z-1$ is the charge number of ions, Z is the atomic number, U_p^k and I_{k+1} are the partition function and the ionization potential energy of the k-fold ion, respectively, and ΔI_{k+1} is the correction term for ionization potential due to the excess free energy, of which the expression is given by

$$\Delta I_{k+1} = \left(\frac{\partial F_C}{\partial N_k}\right)_{T,V} - \left(\frac{\partial F_C}{\partial N_{k+1}}\right)_{T,V} - \left(\frac{\partial F_C}{\partial N_e}\right)_{T,V}. \tag{6}$$

We use the effective charge number defined by

$$Z_{eff}^2 = \overline{z^2} \equiv \sum_{k=1}^{Z} z_k^2 N_k / N_i, \tag{7}$$

to finally obtain

$$\Delta I_{k+1} = -(2k+1)\frac{e^2}{4\pi\epsilon_0 a_i}\left(\frac{\partial f_C^{i\text{-}i}}{\partial \Gamma_i}\right) - \frac{1}{3\bar{z}}\left[\frac{e^2}{4\pi\epsilon_0 a_e}\left(\frac{\partial f_C^{i\text{-}e}}{\partial \Gamma_e}\right)_{r_s} - r_s k_B T\left(\frac{\partial f_C^{i\text{-}e}}{\partial r_s}\right)_{\Gamma_e}\right] - k_B T f_C^{e\text{-}e} - \frac{1}{3}\frac{e^2}{4\pi\epsilon_0 a_e}\left(\frac{\partial f_C^{e\text{-}e}}{\partial \Gamma_e}\right)_{\theta} + \frac{2}{3}\theta k_B T\left(\frac{\partial f_C^{e\text{-}e}}{\partial \theta}\right)_{\Gamma_e}. \tag{8}$$

Now inserting proper forms of the free energy functions and their partial derivatives in the potential correction term, Eq. (8), and solving a full set of coupled equations for ionization balance given by Eq. (5), one can obtain the concentrations of ion and electron species in nonideal plasma. Here, we adopt the analytic formulas presented by Chabrier and Potekhin [12] for the excess free energies associated with the ion-ion and ion-electron interactions denoted by $f_C^{i\text{-}i}$ and $f_C^{i\text{-}e}$, respectively, which accurately fit the results from large-scale Monte Carlo simulations and HNC calculations for fully ionized electron-ion Coulomb plasmas. For electron-electron interactions, an analytic expression $f_C^{e\text{-}e}$ given by Tanaka, Mitake, and Ichimaru [9] is employed, which has been derived from an extensive theoretical study of electron fluids based on the Singwi-Tosi-Land-Sjölander approximation [26].

III. ELECTRICAL CONDUCTIVITY

The electrical conductivity of a plasma medium crucially depends on the degree of ionization: In fully ionized plasma, the motions of current-carrying electrons are governed by Coulomb interactions between electrons and ions while those in weakly ionized plasma are significantly influenced by the head-on collisions between electrons and neutral atoms. A reasonably accurate estimation of the electrical conductivity in partially ionized plasma, σ, can be given by a rather simple model of additive collision frequencies that makes a compromise between the fully and weakly ionized conditions [3,27,28]:

$$\frac{1}{\sigma} = \frac{1}{\sigma_{e\text{-}i}} + \frac{1}{\sigma_{e\text{-}n}}, \tag{9}$$

where $\sigma_{e\text{-}i}$ and $\sigma_{e\text{-}n}$ are the electrical conductivities associated with the electron-ion and electron-neutral collisions, respectively. This formula yields the exact values in both limiting boundaries though there may be uncertainties at an intermediate degree of ionization.

In classical plasmas, $\sigma_{e\text{-}i}$ can be obtained from the Spitzer-Härm formula [29]. This classical plasma conductivity, however, displays a diverging characteristic in strongly coupled conditions because the value of Coulomb logarithm tends to be inaccurate approaching zero as the plasma density increases to a solid level. There are several theoretical models devised to be applicable to nonideal plasmas in a fully ionized regime [30–33]. Among them, the Zollweg-Liebermann model [32], which is based on a more accurate evaluation of the electron-ion collision cross section with the lower limit of the cutoff shielding length defined by the interionic distance, appears more preferable for its simplicity in mathematical expression and reasonable agreement with experimental data and can be readily incorporated with our ionization balance calculation being written as

$$\sigma_{e\text{-}i}=\frac{1}{38}\frac{T^{3/2}}{Z_{eff}}\frac{\gamma_E}{\ln(1+1.4\Lambda_m^2)^{1/2}}, \tag{10}$$

where γ_E is the correction factor for electron-electron collisions, $\Lambda_m=\Lambda[1+(a_i/\lambda_D)^2]^{1/2}$ is the ratio of the modified cutoff shielding length to the impact parameter with the classical definition of Λ [29], and λ_D is the Debye shielding radius.

For calculation of $\sigma_{e\text{-}n}$, we make use of a simple formula

$$\sigma_{e\text{-}n}=\frac{n_e e^2}{m_e \overline{\nu}_{e\text{-}n}}, \tag{11}$$

where $\overline{\nu}_{e\text{-}n}$ is the mean electron-neutral collision frequency. From the first Sonine-polynomial approximation of the exact Chapman-Enskog theory, this collision frequency is given by $\overline{\nu}_{e\text{-}n}=\overline{C}_e n_0 \overline{Q}_{e\text{-}n}$, where $\overline{C}_e=(8k_BT/\pi m_e)^{1/2}$ is the electron mean thermal speed, n_0 is the number density of neutral atoms, and $\overline{Q}_{e\text{-}n}$ is the average momentum transfer cross section of electron-neutral collision [28,34]. Here, we obtain the momentum transfer cross section by using a fitted formula presented by Desjarlais as a function of density and temperature for practical uses in numerical procedures [19]. The resulting values of $\overline{Q}_{e\text{-}n}$ for aluminum atom appear in the range of $(0.2–3.0)\times10^{-18}$ m^2 for temperatures of a few eV and densities of $\rho\leqslant10$ g/cm^3, which is of the same order of magnitude as those found in other papers [17,19].

IV. RESULTS AND DISCUSSION

A. Plasma composition

Calculations are carried out using the atomic energy level data for all ionic charge states of aluminum [35] without considering the electronic excitation. Figure 1 shows the profile of electron concentration $\alpha_e=n_e/(n_0+n_i)$ in aluminum plasma in a two-dimensional parameter space of density $\rho=(10^{-5}–10)$ g/cm^3 and temperature $T=(10^4–10^7)$ K obtained by solving the coupled equations of ionization balance through fully iterative numerical procedures with the corrected ionization potentials. The electron concentration, here, exhibits a behavior of strong dependence on temperature rather than on density in the density range far below the solid density level, i.e., $\rho<1$ g/cm^3. The stepwise shape of this profile along the temperature axis reveals the electronic configuration of aluminum atom. At higher densities, on the other hand, the Coulomb coupling effect results in an unusual pattern in the electron concentration profile. A sudden increase of the degree of ionization occurs near the solid density at low temperatures $T<10^5$ K, which, we consider, effectively demonstrates the physical situation referred to as the pressure ionization that eventually leads to the metal-insulator transition.

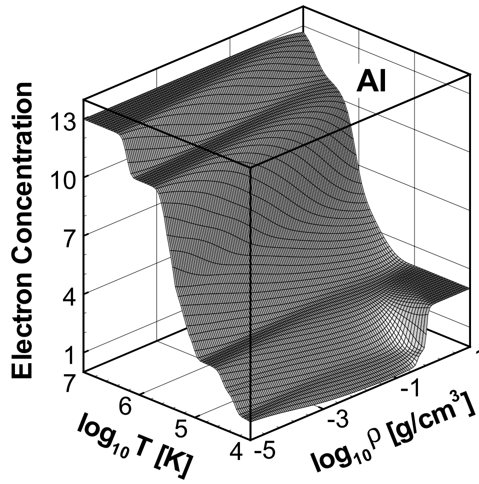

FIG. 1. Electron concentration in aluminum plasma obtained from fully iterative calculations using the present ionization balance model in a density-temperature space.

The detailed aluminum ion concentrations are presented in Fig. 2 for various charge states at different temperatures of (a) 10 000 K, (b) 14 000 K, (c) 20 000 K, and (d) 31 600 K. Here, the maximum value of ion charge number is found to be $+3$ under the regime of pressure ionization in the density range of $\rho=(1–10)$ g/cm^3. Therefore, the assumption of limited charge number up to $+3$ used in the previous calculation by Redmer [24] seems acceptable at least for aluminum plasma within this physical parameter range. Our results appear overall consistent with those of Redmer's except for the case of $T=10\,000$ K, see Fig. 2(a), where we have the maximum charge number of $+3$ instead of $+2$. In Figs. 2(b)–2(d), it is shown that the sharply rising curve of the electron concentration at $\rho\sim1$ g/cm^3 becomes smoother as the temperature increases because the relative strength of Coulomb coupling is gradually reduced; and further, for much higher temperatures, i.e., $T>10^5$ K, as shown in Fig. 1, the density effect is substantially overridden by the thermal effect so that the contribution by pressure ionization is hardly distinguishable within the density range considered here.

B. Electrical conductivity

Figure 3 shows the electrical conductivities of aluminum plasma calculated by a number of theoretical models using the plasma compositions obtained above in comparison with data from recent experiments measured by Mostovych and Chan [13], DeSilva and Katsouros [14], Krisch and Kunze [15], and Benage, Shanahan, and Murillo [16,17]. In this figure, the dotted and dashed lines represent the electrical conductivities given by Spitzer and Härm [29] and Zollweg and Liebermann [32], respectively, with no neutral effect in-

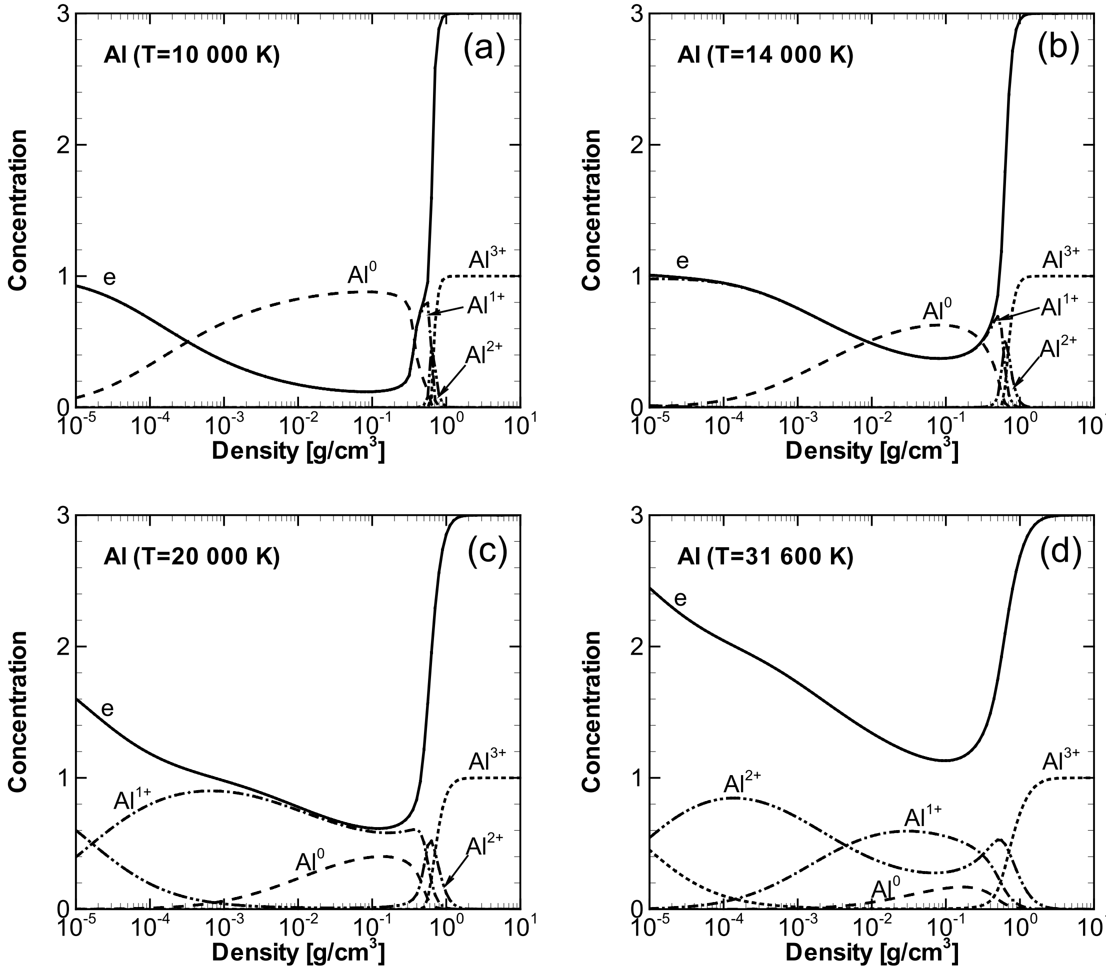

FIG. 2. Electron and ion concentrations in aluminum plasma computed for different temperatures of (a) 10 000 K, (b) 14 000 K, (c) 20 000 K, and (d) 31 600 K as a function of density.

cluded under the assumption of fully ionized plasma whereas the solid lines are computed in a partially ionized plasma regime by a linear mixture rule of additive collision frequencies, Eq. (9), which takes into account both the electron-ion and electron-neutral collisions by incorporating the Zollweg-Liebermann conductivity [32] with the electron-neutral momentum transfer cross section of Desjarlais [19].

The Spitzer-Härm conductivities appear with reasonable values in the low-density limit but tend to become uncontrollably larger as the density increases. On the other hand, the Zollweg-Liebermann model which takes into account the dense plasma effect yields finite conductivities at high densities and seems to agree well with the measurements made by Benage, Shanahan, and Murillo in the upper regime of pressure ionization where plasma is fully ionized. For sufficiently low densities, the Zollweg-Liebermann conductivity well recovers the Spitzer-Härm value. However, the Zollweg-Liebermann model neglecting the electron-neutral collisions is unable to describe the conductivity behavior in the physical range where a considerable fraction of neutral atoms exist.

With the neutral effect taken into account, Eq. (9) yields remarkably decreased values of electrical conductivity below the critical density at which the pressure ionization occurs, particularly, for low temperatures where neutral particles dominate. In Fig. 3(a), for example, the partially ionized plasma conductivity computed for $T = 10\,000$ K has its local minimum at $\rho \sim 0.1$ g/cm^3 with the minimum value found to be lower than that predicted by the Zollweg-Liebermann model by about an order of magnitude. Compared with the measured data of DeSilva and Katsouros at the same temperature, the present mixture-rule calculation seems to overestimate the neutrals' contribution leading to a steeper slope of the conductivity-density curve but, nevertheless, successfully reproduces the metal-insulator transition. At $T = 31\,600$ K, on the contrary, see Fig. 3(d), due to the vanishing population of neutral particles as a result of thermal ionization, the electrical conductivities calculated by Eq. (9) for partially ionized plasma become very close to those by the fully ionized model given by Zollweg and Liebermann over the entire density range. This signifies that it may be sufficient to use a conductivity model that takes account only of the electron-ion collisions for temperatures $T \geqslant 30\,000$ K in the density range of $\rho \leqslant 10$ g/cm^3 reducing the computational effort.

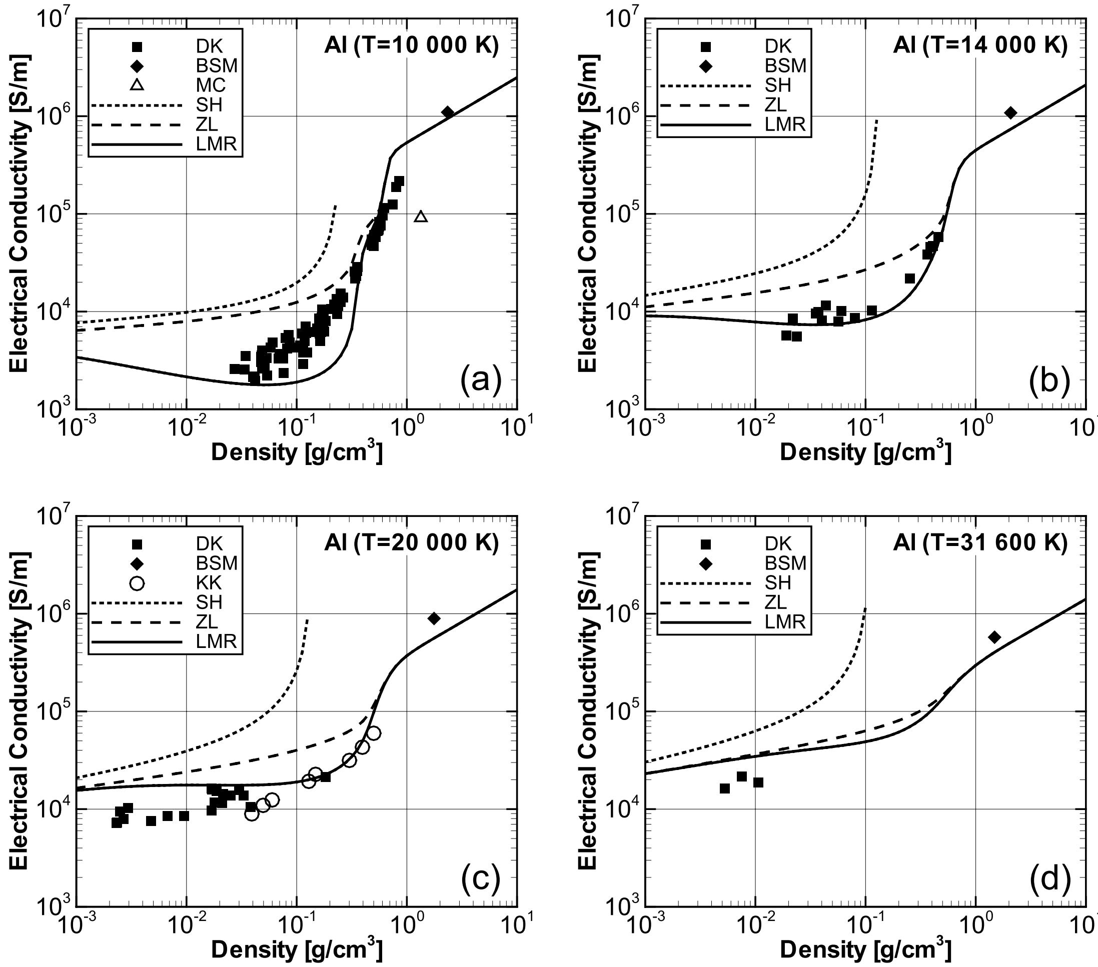

FIG. 3. Electrical conductivity of aluminum plasma computed by the present partially ionized plasma model based on a linear mixture rule (LMR) for different temperatures of (a) 10 000 K, (b) 14 000 K, (c) 20 000 K, and (d) 31 600 K as a function of mass density in comparison with the experimental data measured by DeSilva and Katsouros (DK), Benage, Shanahan, and Murillo (BSM), Mostovych and Chan (MC), and Krisch and Kunze (KK) along with the theoretical predictions for fully ionized plasma given by Spitzer and Härm (SH) and Zollweg and Liebermann (ZL).

Discrepancies between calculations and experiments can be attributed to the fact that the laboratory plasma is in most cases away from the equilibrium state if the density is low and some experiments resort to their own equation-of-state models for data interpretations, aside from the inherent inaccuracy in Eq. (9). Nevertheless, the present calculation seems to well reproduce the general trend of measured data over a significant physical range supporting the validity of the theoretical models employed in this work for the excess free energy and the electrical conductivity in a practical aspect. Further, we recognize that it would effectively enhance the reliability of calculated results to implement a more realistic model for electron collision processes in partially ionized plasmas instead of using a simple linear mixture rule [24,28,36].

V. SUMMARY

In this paper, we calculated the electrical conductivity of aluminum plasma in a wide density-temperature range covering the strongly coupled regime. The correction term for ionization potential was formulated using the dimensionless parameters characterizing nonideal plasmas. A set of fitted formulas of excess free energy, which have been derived from extensive theoretical studies and massive numerical procedures, were utilized to determine the ionization balance. The evaluation of electrical conductivity in a partially ionized condition is based on a linear mixture rule of additive collision frequencies for electron-ion and electron-neutral interactions. The calculated plasma compositions appear to well demonstrate the pressure ionization occurring near the solid density at temperatures of a few eV. The resulting electrical conductivity profiles also reproduce the overall trend of recently measured data to a substantial degree. From the results, it follows that the present physical models for the corrected ionization balance and the electrical conductivity in nonideal plasma are reasonably acceptable in depicting the metal-insulator transition.

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