Semi-empirical model for permittivity of warm dense matter

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Abstract. The permittivity of Lorentz plasmas and wide-range expression for effective frequency of collisions of electrons are considered in wide temperature range. The proposed expression fulfills all limiting cases (high- and low-frequency skin effect, degenerate and non-degenerate plasmas) and permits one to incorporate different physical phenomena in warm dense matter like contribution of electron-phonon collisions and umklamp process to permittivity. For the case of aluminum plasmas, the model takes into account both intraband and interband contributions to permittivity via semi-empirical Huttner model modified in such a way to ensure proper description of optical properties of aluminum at room temperatures.

1. Introduction
Consequent description of laser or particle beams interaction with matter requires information on equations of states [1, 2] as well as transport properties [3] and optical properties of warm dense matter (WDM). To describe optical properties, information on permittivity \( \varepsilon \) of matter is necessary.

Particularly, the information on \( \varepsilon \) is required for simulations of electron [4] and ion [5, 6] stopping power and determination of the rate of laser energy absorption [7–14] in matter.

Until now rather sophisticated methods are elaborated for calculation of \( \varepsilon \), including fundamental quantum statistical (QS) approach within linear response theory [15–17] and first-principle DFT-MD calculations [18–20] based on use of Kubo–Greenwood formula [21]. Those approaches, though they are based on the fundamental theoretical principles, have their own drawbacks. Particularly, QS approach give rise to final expressions for \( \varepsilon \), generally speaking, only in the case of moderate or weak coupling, when electron-ion coupling parameter \( \Gamma \lesssim 1 \) and one can allocate some small parameter of interaction to implement Green’s function technique and construct the theory [15–17]. DFT-MD approach due to numerical difficulties is presently used only for relatively moderate temperatures of the order of several eV or lower. Besides, DFT-MD calculations of \( \varepsilon \) are too time consuming to be used in hydrodynamical codes.

Therefore semi-empirical models which are based, from one hand, on known theoretical principles in the region of weakly coupled plasmas, and from another hand, on heuristic analysis and experimental data in the region, where purely theoretical considerations are difficult, are actual. Such a model is proposed in the present paper.
2. Formulation of the model

The permittivity of WDM can be expressed as

\[ \varepsilon = \varepsilon_{bb} + 1 - (n/n_c)m_{opt}^{-1}K_0, \]  

where \( \varepsilon_{bb} \) is interband part of permittivity [22] and another terms in (1) stands for Drude-like or intraband part of permittivity, where \( n \) and \( n_c = m_\omega^2/(4\pi e^2) \) are electrons concentration and critical concentratoin, respectively, \( m \) and \( e \) are electron mass and charge, respectively, \( \omega \) is laser frequency; \( m_{opt} \) is optical mass of electron.

From the Boltzmann kinetic equation in \( \tau \)-approximation, the expression for the value of \( K_0(\omega) \) in the formula (1) can be written as

\[ K_0(\omega) = \frac{i \omega m}{n} \int \frac{v^2}{3} \frac{2d^3p}{(2\pi \hbar)^3} \frac{\partial f_F}{\partial \epsilon} \frac{1}{\nu(\nu - i\omega)}, \nu = \frac{4\pi ne^4\Lambda Z}{m^2v^4}, \]  

where \( f_F \) is Fermi distribution of electrons over energies \( \epsilon = p^2/(2m) = mv^2/2 \), \( \Lambda \) is Coulomb logarithm, which, generally speaking, is both energy [23, 24] and frequency [25–28] dependent.

After a dimensionless transformation, and taking derivative of Fermi function this formula can be rewritten as

\[ K_0(\omega) = e^{-3/2} \int_0^\infty \frac{e^{\xi - \epsilon_F}}{[1 + e^{\xi - \epsilon_F}]} \frac{\xi^{3/2} - i\nu_T(\xi, \omega)}{\xi^3 + \nu_T^2(\xi, \omega)} \xi^3 d\xi, \]  

with

\[ \xi = \epsilon/T, \nu_T = (3\sqrt{\pi}/4)\nu_{pl}/\omega, \]

where \( \epsilon_F = E_F/T, \epsilon_\mu = \mu/T; \mu, E_F = h^2/(2m)(3\pi^2n)^{2/3} \) and \( T \) are chemical potential, Fermi energy and electrons temperature, respectively; \( \nu_{pl} \) is electron-ion frequency of collisions:

\[ \nu_{pl}(\xi, \omega) = \frac{4\sqrt{2\pi}}{3} \frac{ne^4Z}{\sqrt{mT^{3/2}}} \Lambda(\xi, \omega), \]

where \( Z = n/n_i \) is average ions charge, \( n_i \) is total concentration of heavy particles.

Taking in mind the limiting cases derived below, it is reasonable to introduce the effective frequency of collisions in the following way:

\[ \nu_{eff} \triangleq \epsilon_F^{-3/2}[1 + e^{-\epsilon_F}]-1\nu_T. \]  

The above expressions were rigorously derived for the case of a weakly coupled plasma. Further we postulate their applicability for the case of a strongly coupled plasma, but with replacement of \( \nu_{eff} \) by the value, determined by the procedure described below. In this case the equality (5), which serves above as the definition of \( \nu_{eff} \), below is taken – reversely – as a definition of the value of \( \nu_T(\omega) \) in the integral (3):

\[ \nu_T(\xi, \omega, \epsilon_F) = (\nu_{eff}(\xi, \omega)/\omega)\epsilon_F^{-3/2}[1 + e^{-\epsilon_F}]. \]  

One can use different models for effective frequency of collisions \( \nu_{eff} \) (see, for example, [7, 8, 29, 30]). In any case, in accordance with phisical processes involved, \( \nu_{eff} \) in wide density and temperature range can be expressed via some interpolation between effective frequency of collisions in dense matter at low temperatures with metallic-like behavior \( \nu_{met} \), maximum possible frequency of collisions in strongly coupled plasmas at intermediate temperatures
\(\nu_{\text{max}} [7,23]\) and classical plasmas frequency of collisions \(\nu_{pl}\) at high temperatures, like in the form proposed in [8]:

\[
\nu_{\text{eff}}(\xi, \omega) = \nu_{\text{max}}[1 + (\nu_{\text{max}}/\nu_{\text{met}})^6 + (\nu_{\text{max}}/\nu_{pl})^6]^{-1/6},
\]

where

\[
\nu_{\text{max}} = k_1 \omega_{pl}
\]

and

\[
\nu_{\text{met}} = \nu_{dc} + \beta_\nu(T_i - T_{\text{room}}) + k_2 T_e^2/(\hbar E_F),
\]

where \(T_i\) is ions temperature and \(T_{room}\) is room temperature (generally speaking, \(T \neq T_i\)).

Coefficients \(k_1 \lesssim 1\) and \(k_2 \lesssim 1\) in (9) and (8) can be found from a comparison of calculated results with the experimental data [8,31]; 1-st & 2-nd terms in (9) can be determined as [32]

\[
\nu_{dc} = \nu_{\text{dc}0}\omega_p/(4\pi m_{opt}), \quad \beta_\nu = \beta_\nu\omega_p/(4\pi m_{opt}),
\]

where \(\omega_p = \sqrt{4\pi ne^2/m}\) is plasma frequency and coefficients \(\nu_{\text{dc}0}\) and \(\beta_\nu\) are determined from measurements of stationary conductivity.

Taking in mind that energy dependence in Coulomb logarithm is slower than for other terms under integral in (3), one can replace in (4) \(\Lambda(\xi, \omega)\) by it’s value at some \(\xi = \xi_*\), i.e. use \(\Lambda\) at some fixed energy (similarly as it was done in [33]).

Below the model similar to modified Spitzer-Harm model [33] for \(\Lambda\) is used, with the exception that for relatively high laser frequencies correction of the screening by the value of \(v_{th}/\nu_{\text{eff}}\) [25–28] (\(v_{th} = \sqrt{T/m}\)) is taken into account:

\[
\Lambda = \frac{1}{2} \ln \left[ 1 + \left( \frac{3}{\Gamma} \right)^2 \min \left\{ 1, \frac{\omega_p^2}{\omega^2} \right\} \right], \quad \tilde{\Gamma} = \frac{Ze^2}{\max\{\lambda_{DH}, R_0\}},
\]

where \(\lambda_{DH} = v_{th}/\omega_p\left[1 + \frac{\omega_p^2}{\omega^2}\right]^{1/2} + ZT/T_i\right]^{-1/2}\) is Debye-Huckel screening length [34], \(R_0 = (4\pi n_i/3)^{-1/3}\) is interatomic distance [34].

3. Limiting cases

The above model fulfills all known limiting cases where it gives rise to well-known expressions:

(i) Drude case \(T \ll E_F\):

\[K_0 = [1 + i(\nu_{\text{eff}}/\omega)]^{-1}, \quad \nu_T = (\nu_{\text{eff}}/\omega)(E_F/T)^{3/2}.\]

(ii) Non-degenerate plasma, \(E_F \ll T\):

\[K_0 = K_1(\nu_T) - i(\nu_{\text{eff}}/\omega)K_2(\nu_T), \quad \nu_T = (3/4)\sqrt{\pi}(\nu_{\text{eff}}/\omega),\]

\[K_1(x) = \frac{8}{\sqrt{\pi}} \int_0^\infty e^{-t^2} t^6(1+x^2)^{-1} dt, \quad K_2(x) = 2 \int_0^\infty e^{-t^2} t^7(1+x^2)^{-1} dt, \quad \text{see, e.g. [8,35,36]}.\]

Asymptotics of \(K_1\) and \(K_2\): for \(\nu_T \to 0\): \(K_1 \to 1, K_2 \to 1\); for \(\nu_T \to \infty\): \(K_1 \to 70/\pi(\nu_{\text{eff}}/\omega)^{-2}, K_2 \to 32/(3\pi)(\nu_{\text{eff}}/\omega)^{-2}\).

(iii) High frequencies, \(\omega \gg \nu_{\text{eff}}\):

\[K_0 = 1 - i\nu_{\text{eff}}/\omega.\]

(iv) Low frequencies, \(\omega \ll \nu_{\text{eff}}\):

\[K_0 = \frac{B_1(\epsilon_{\mu})}{(\nu_{\text{eff}}/\omega)^6} - i \frac{B_2(\epsilon_{\mu})}{\nu_{\text{eff}}/\omega},\]

\[B_1(\epsilon_{\mu}) = \frac{4}{3} F_2(\epsilon_{\mu}) F_{1/2}(\epsilon_{\mu})[1 + e^{-\epsilon_{\mu}}]^{-2}, \quad B_2(\epsilon_{\mu}) = \frac{4}{3} F_2(\epsilon_{\mu}) F_{1/2}(\epsilon_{\mu})[1 + e^{-\epsilon_{\mu}}]^{-1}.\]

Asymptotics of \(B_1\) and \(B_2\): for \(E_F \ll T\): \(B_1 \to 70/\pi, B_2 \to 32/(3\pi), \) for \(E_F \gg T\): \(B_1 \to 1, B_2 \to 1\).
4. Interband conductivity for aluminum plasmas

In this section semi-empirical model for interband part of conductivity \( \varepsilon_{bb} \) for aluminum plasmas is given.

In accordance with [32], the permittivity \( \varepsilon_{bb} \) can be written as

\[
\varepsilon_{bb} = 1 - \frac{n_e}{n_c} \sum_n \frac{A_n}{(\omega_{bbn}/\omega)^2 - (1 + i\nu_{bb}/\omega)^2} \tag{12}
\]

where \( A_n \) is an average oscillator strength, \( \omega_{bbn} \) are resonance frequencies, which correspond to transitions between parallel bands for different directions in reverse \( k \)-space. For aluminum there are two kind of transitions: in the directions with crystal indexes 111 and 200, with frequencies \( \omega_{bb1} = 2U_{111}/h \) and \( \omega_{bb2} = 2U_{200}/h \), where \( U_{111} \) and \( 2U_{200} \) are Fourier-components of pseudo-potential at respective directions, \( \hbar \omega_{bb1} = 0.457 \text{ eV} \) and \( \hbar \omega_{bb2} = 1.53 \text{ eV} \) at room temperature; \( \nu_{bb} \) is characteristic frequency of collisions, responsible for dumping of interband absorption. In the case of aluminum the values of \( A_n \), \( n = 1, 2 \) can be determined, in accordance with [32], from optical theorem

\[
\sum_n A_n = 1 - 1/m_{opt}, \tag{13}
\]

where \( m_{opt} = m_{e,\text{dens}}/m_e \) is optical mass of an electron (\( m_{e,\text{dens}} \) is an effective mass of an electron in condense matter), and from another equality, which follows from (12) for aluminum, if one take into account, that \( \nu_{bb}/\omega \ll 1 \), \( n = 1, 2 \):

\[
\frac{A_1}{A_2} = \frac{\omega_{bb1} \nu_{bb} (\omega_{bb1})}{[\omega_{bb2} \nu_{bb} (\omega_{bb2})]} \frac{\nu_{bb} (\omega_{bb1})}{\nu_{bb} (\omega_{bb2})} \tag{14}
\]

The value of \( \varepsilon \) at room temperature can be determined from hand-book room-temperature optical constants \( n = n(\omega) \) and \( k = k(\omega) \), in accordance with the definition \( \varepsilon = (n + ik)^2 \) \((n \text{ and } k \text{ are real values})\). Taking in mind that at room temperatures \( T_e = T_i = T_{room} \) the Drude-like formula is applicable for \( \varepsilon_D \),

\[
\varepsilon_D = 1 - (n_e/n_c)m_{opt}^{-1}(1 + i\nu_{eff}/\omega)^{-1}, \tag{15}
\]

where \( \nu_{eff} \) is effective frequency of electron-ion (electron-phonon) collisions, one can derive from (14) the following expression for \( A_1/A_2 \):

\[
\frac{A_1}{A_2} = \frac{\omega_{bb1} \nu_{bb} (\omega_{bb1})}{\omega_{bb2} \nu_{bb} (\omega_{bb2})} \left[ \frac{2n_1 k_1 - (\omega_{pl}/\omega_{bb1})^2 m_{opt}^{-1}(\nu_{eff}/\omega_{bb1})}{2n_2 k_2 - (\omega_{pl}/\omega_{bb2})^2 m_{opt}^{-1}(\nu_{eff}/\omega_{bb2})} \right]. \tag{16}
\]

where \( n_l = n(\omega_{bb})k_l = k(\omega_{bb}), \ l = 1, 2 \); room-temperature values are assumed in (16). From (13) and (16) the expressions for \( A_1 \) and \( A_2 \) are the following:

\[
A_1 = B(1 - m_{opt}^{-1})/(1 + B), \quad A_2 = (1 - m_{opt}^{-1})/(1 + B), \tag{17}
\]

where \( B \equiv A_1/A_2 \).

The values of \( m_{opt}, \omega_{bbn} \) and \( \nu_{bb} \) in (12) are, generally speaking, depend on ion temperature \( T_i \) [32]. Taking in mind [32], the temperature and density dependence of \( m_{opt} \) and \( \omega_{bbn} \) can be approximated as

\[
m_{opt}(T_i) = 1 + [m_{opt0} - 1][1 + \beta(T_i - T_{room})^{-1}]\theta(\rho - \rho_{dm}),
\]

\[
\omega_{bbn}(T_i) = \omega_{bbn,0}[1 + \beta(T_i - T_{room})^{-1}]\theta(\rho - \rho_{dm}), \tag{18}
\]

\[\]
Figure 1. Dependence of absorption coefficient (a), $\text{Re}\{\varepsilon\}$ (b), $\text{Im}\{\varepsilon_{bb}\}$ (c) and $\text{Im}\{\varepsilon\}$ (d) on photon energy $\hbar\omega$ for $T_e = T_i = 0.025, 0.1$ and $150$ eV (solid, dashed and dotted lines, respectively), thin lines for $\alpha_0 = 4$, $\alpha_1 = \alpha_2 = 0$ in (21), thick lines for $\alpha_0 = 0.3$, $\alpha_1 = 1.7$, $\alpha_2 = 0.1$. Markers are from textbook of Palik [37] for room-temperature values.

where $\theta$ is Heviside function and $\rho_{dm}$ is the characteristic density of dense matter, which can be chosen as minimum between liquid and solid state density; $m_{\text{opt}}$, $\beta$ and $\omega_{bb,0}$ are constants. For aluminum, in accordance with [32], they are equal to:

$$
\omega_{bb1,0} = \omega_{bb1}(T_{\text{room}}) = 0.457 \text{ eV}/\hbar,
\omega_{bb2,0} = \omega_{bb2}(T_{\text{room}}) = 1.53 \text{ eV}/\hbar;
$$
$$
m_{\text{opt}} = \begin{cases} 1.5, & \text{solid matter}, \\ 1.28, & \text{liquid matter}; \end{cases}
$$
$$
\beta = \begin{cases} 7.5 \times 10^{-5}K^{-1}, & \text{solid matter}, \\ 1.5 \times 10^{-4}K^{-1}, & \text{liquid matter}. \end{cases}
$$

The expression for $\nu_{bb}$ can be written in the form, similar to (9) (but without contribution from electron-electron collisions):

$$
\nu_{bb} = \alpha_w \nu_{dc} + \beta \nu(T_i - T_{\text{room}}), \quad \alpha_w = (\alpha_0 + \alpha_1 \omega^2)/(1 + \alpha_2 \omega^2),
$$

where, unlike the similar expression in [32], where $\alpha_w = \text{const} = 4$, the phenomenological dependence on $\omega$ through the term $\alpha_w$ is introduced in such a way, to reproduce the experimental dependence of room-temperature value of absorption coefficient $A$ on $\omega$, see figure 1.

From above equations for $\varepsilon_{bb}$ and figure 1 one can see that at ion temperatures ($T_i \gtrsim 1$ eV) much above the melting temperature interband contribution to imaginary part of permittivity of aluminum at optical frequencies (two peaks at figure 1(c)) vanishes due to smearing of the respective parallel bands structure. Nevertheless, due to the integral Kramers-Kroning
Figure 2. Dependence of real (a) and imaginary (b) parts of permittivity and absorption coefficient (c) on temperature $T = T_i$ of solid-density aluminum plasmas: solid curves for present model, dashed curves for model of [29] and dotted curve for model of [39].

relations all possible interband transitions in entire frequency domain can produce an essential contribution to real part of permittivity even at higher temperatures [38].

5. Comparison with other models and conclusion
In figure 2 the model described above is compared with the models proposed earlier in [39] and [29]. The later model had used free parameters determined from simulation of experimental data on phase change and reflectivity of S- and P-polarized femtosecond laser probe pulses reflected from hot solid aluminum target heated by pump femtosecond laser [40].

Both present model and the model of [29] gives higher peak values of absorption coefficient, than earlier model [39]. So the model [39] underestimates absorption at relatively low temperatures $T \leq 40$ eV (and overestimates it for $T > 40$ eV).

The positions of peaks are slightly different for the present model and [29]. This difference is mainly due to different description of electron-phonon interaction part of the effective collision frequency (in the present model the result proposed in [32] is used).

The advantage of the present model in comparison with that described in [29] is that it proposes consequent derivation of the interpolation expression for permittivity in wide temperature range, which fulfills known limiting cases. Also it gives more consequent description of the interband contribution to permittivity. The model formulated above permits one to describe optical properties of matter in wide range of plasma parameters. For such description
one should use information about constants $n_{1,2}$, $k_{1,2}$, $m_{\text{opto}}$, $\theta_{\text{dc}}$, $\beta$, and (for aluminum) $\omega_{\text{obj}}, \beta_\nu$, which can be taken from handbooks or independent experiments, and use fitting parameters $k_{1,2}$ and (for aluminum) $\alpha_{0,1,2}$, which can be determined, for example, from data on measurements of optical properties of laser heated matter [8, 29, 31].

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