Influence of optical non-uniformity on the reflectance of dense plasmas

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Abstract. We provide theoretical analysis of the reflectance of shock compressed plasmas and warm dense matter for normal incidence of laser radiation as well as for the dependence of s- and p-polarized reflectivity on incidence angle. We use density functional theory approach for the calculation of the dielectric function and reflectivity. The Kohn–Sham set of equations with the projector augmented wave (PAW) potential is solved for valent electrons. Due to the nonlocality of the PAW potentials, the longitudinal expression for the imaginary dielectric function is used. The real part is obtained by the Kramers–Kronig transformation. Quantum molecular dynamics simulation and VASP is used. Comparison with the experimental data for shock compressed xenon is performed. Three wavelengths are considered.

1. Introduction
The reflectance of the shock-compressed xenon is measured in the experiments of Mintsev and Zaporozhets [1–4]. There are a lot of theoretical attempts to describe these experimental data. Taking into account the static collisional frequency within the framework of the Drude model [5] does not give a satisfactory explanation of the results obtained. The more accurate expressions for the dynamic collisional frequency in the Born approximation [5] do not improve agreement with the experiment. Other attempts to explain experimental results within the framework of the Drude model are associated with an artificial broadening of the wavefront [3–8]. Despite a certain improvement of the agreement with the experiment, this approach does not allow to establish one-to-one correspondence between the reflectance values and the free charge concentration. Moreover, the agreement with the experiment for normal incidence of laser radiation is reached, if the shock front width is 800 nm, which is much higher than the initial theoretical estimation 100 nm [1] (the estimation is based on evaluation of the xenon ionization rate). The value of the width 220 nm gives satisfactory agreement of theoretical results with the experiment for the dependence of polarized reflectivity on incident angle [4]. However, this value of the width makes worse the agreement with the experiment for normal reflectance.

The approach for the self-consistent description of optical and electronic properties of nonideal plasmas and warm dense matter is suggested in [9–13]. The approach gives satisfactory description of experiments on normal reflectance of xenon plasmas [9]. The approach is applied for description of electronic properties of warm dense hydrogen and selenium in the region of the phase transition [12].

In this work, the approach is applied for description of the reflectance from the broadened shock wave front in xenon plasmas. In section 2, the method of calculation of the dielectric
function (DF) and reflectivity within the framework of the density-functional theory (DFT) is discussed. The case of the reflection from the ideal shock-wave front is considered. The idea of the self-consistent approach for description of optical and electronic properties of warm dense matter and nonideal plasmas is briefly described. The methods of calculation of reflectivity from the non-uniform density profile and comparison with the experimental data [1–4] for xenon plasma are considered in section 3. The cases of normal incidence of laser radiation (subsection 3.1) and s- and p-polarized reflectivity (subsection 3.1) are discussed. Experimental results for dependence of s- and p-polarized reflectivity on incident angle [4] are used to investigate the influence of the optical non-uniformity to the optical properties of xenon plasmas. There are conclusions in section 4.

2. Self-consistent calculation method

The Fresnel formulas are used for calculation of reflectivity for the normal incidence of the electromagnetic wave

\[ R = \left| \frac{(\sqrt{\varepsilon} - 1)}{(\sqrt{\varepsilon} + 1)} \right|^2, \quad (1) \]

and for the s- and p-polarized reflectivities

\[ R_s = \frac{(\cos \varphi - \sqrt{\varepsilon - \sin^2 \varphi})^2}{(\cos \varphi + \sqrt{\varepsilon - \sin^2 \varphi})^2}, \quad R_p = \frac{(\varepsilon \cos \varphi - \sqrt{\varepsilon - \sin^2 \varphi})^2}{(\varepsilon \cos \varphi + \sqrt{\varepsilon - \sin^2 \varphi})^2}. \quad (2) \]

The DF, which is included in (2) and (1) is a complex function and can be expressed as \( \varepsilon = \varepsilon^{(1)} + i \cdot \varepsilon^{(2)} \). We consider the interaction of the electromagnetic (transverse) radiation with matter and the response function is the transverse DF. The dependence on frequency \( \omega \) of the imaginary part of the transverse DF is defined by the following expression in the long-wavelength limit

\[ \varepsilon^{(2)}(\omega, \mathbf{R}_1) = (4\pi^2 e^2 / 3\omega^2 \Omega) \lim_{|q| \rightarrow 0} \sum_{n,n',\alpha,\mathbf{k}} 2w_{\mathbf{k}} f(T, E_{n,\mathbf{k}}) - f(T, E_{n',\mathbf{k}+\mathbf{q}}) \times |\langle \psi_{n'\mathbf{k}} | \hat{v}_\alpha | \psi_{n\mathbf{k}} \rangle|^2 \cdot \delta(E_{n',\mathbf{k}+\mathbf{q}} - E_{n\mathbf{k}} - \hbar \omega) \quad (3) \]

at a given ion configuration \( \mathbf{R}_1 \) and temperature \( T \), where \( e \) is the elementary charge, \( \Omega \) is a system volume, \( \mathbf{q} \) is a wave vector of the incident radiation, \( \hbar \) is the Plank constant. The summation is carried out over all electron states \( n, n' \). The summation over index \( \alpha \) multiplied by 1/3 stands for the averaging over three spatial coordinates. The summation is also carried out over all \( \mathbf{k} \)-points in the Brillouin zone taking into account the weights \( w_{\mathbf{k}} \) of the \( \mathbf{k} \)-points. The factor 2 before the weights allows for the electron spin-degeneracy. \( f(T, E_{n,\mathbf{k}}) \) is the Fermi-Dirac distribution function. \( E_{n,\mathbf{k}} \) is an eigenvalue (an energy level) corresponding to the wave function \( \psi_{n,\mathbf{k}} \). \( \psi_{n,\mathbf{k}} \) is a solution of the Kohn–Sham equation. We find this solution as a sum of plane waves and therefore it can be represented by means of the Bloch function \( \psi_{n,\mathbf{k}} = e^{i\mathbf{k} \cdot \mathbf{r}} \cdot u_{n,\mathbf{k}} \), where \( u_{n,\mathbf{k}} \) is a cell periodic part. \( \hat{v} \) is the velocity operator.

For local potentials \( V(\mathbf{r}) \), the expression for velocity operator is equivalent to the momentum operator \( \hat{v} = \mathbf{p} / m \), where \( m \) is the electron mass. Substitution of this result in (3) gives the Kubo–Greenwood formula [14,15]. For the nonlocal potentials \( V(\mathbf{r}, \mathbf{r}') \), the additional term arises in the expression for the velocity operator \( \hat{v} \):

\[ \hat{v} = \mathbf{p} / m + (i / \hbar) \left[ V(\mathbf{r}, \mathbf{r}') , \mathbf{r} \right], \quad (4) \]

since the operators of the potential \( V(\mathbf{r}, \mathbf{r}') \) and distance \( \mathbf{r} \) do not commute. Consequently, the Kubo–Greenwood formula is incorrect for the nonlocal potentials.
The longitudinal and transverse DFs are equal to each other in the long-wavelength limit. In this limit the velocity operator can be expressed as [16]

\[ \hat{v} = \lim_{|q| \to 0} \left[ (p^2/2m + V(r, r')) \exp(iqr) \right] /\hbar |q|. \] (5)

The substitution of (5) in (3) gives the expression for the imaginary part of the longitudinal DF [17–19]:

\[ \varepsilon^{(2)}_L(\omega, R_I) = \frac{4\pi^2k^2}{16} \lim_{|q| \to 0} \frac{1}{|q|} \sum_{n,n',\alpha,k} 2\omega_k \left[ f(T, E_{n,k}) - f(T, E_{n',k+q}) \right] \times \left| \langle u_{n',k+e_{\alpha}q} | u_{n,k} \rangle \right|^2 \delta(E_{n',k+q} - E_{n,k} - \hbar\omega), \] (6)

where the unit vector \( e_\alpha \) determines the direction of the Cartesian axis corresponding to the coordinate \( \alpha \). Since we use the conversion of the velocity operator for the derivation of (6), the longitudinal expression has no disadvantages, which the Kubo–Greenwood formula has. The formula (6) can be used for any electron-ion potentials. The Kubo–Greenwood formula can be used only with corrections, which take into account non-locality of a PAW potential.

The eigenvalues and the wave functions are calculated within the framework of the Kohn–Sham DFT approach. VASP (Vienna ab initio simulation package) [20–23] plane-wave code is used in this work for DFT modeling. The type of the exchange-correlation functional is PBE [24]. It is shown in [25, 26] that the longitudinal expression (6) gives more correct result in comparison with the widely used Kubo–Greenwood formula within the framework of the projector augmented wave (PAW) approach. The correctness of the expression (6) is confirmed in [9, 27] where it is shown that using of (6) provides better explanation of the experimental dependence [1,2,8] of the shocked xenon reflectivity for normal incidence on density in comparison with the Kubo–Greenwood formalism.

The real part of the DF is obtained by the Kramers–Kronig transformation

\[ \varepsilon^{(1)}(\omega, R_I) = 1 + \frac{2}{\pi} \int_0^\infty \frac{d\omega'}{(\omega')^2 - (\omega - \imath\eta)^2} \left( \omega' \varepsilon^{(2)}(\omega', R_I) \right), \] (7)

where \( P \) denotes the principle value (in the limit \( \eta \to 0 \)). It should be noted, that expressions (3), (6) for the DF are obtained for the limit of zero effective collisional frequency \( (\eta \to 0) \) and are applicable for frequencies \( \omega \gg \eta \). However, in nonideal plasmas the frequency of collisions has finite value. The estimation of the influence of the collisions in plasmas to the optical properties is discussed in [5–7, 28–30]. This problem requires separate more detailed consideration, which is out of scope of the present work.

The approach of calculation of the plasma frequency and effective free electron density based on the using of the sum rule is suggested in [9]. The imaginary part of the DF is included in the expressions for both the reflectivity and the plasma frequency. One and the same sum over states defines explicitly both values. Therefore, the values of the plasma frequency calculated within the framework of the approach are directly associated with the dependencies of the reflectivity on the plasma density. It allows to provide an approach for self-consistent description of optical and electronic properties. Optical properties include reflectance, Brewster angle, absorption, transmission. Corrections are considered which allow for the finite width of the transient layer at the WDM border. Electronic properties are conductivity, plasma frequency and electronic density of states. The scheme of the approach is shown in figure 1. In this work, the upper part of the scheme (optical properties) is considered.
3. Reflectance from the non-uniform density profile

3.1. Normal incidence

For normal incidence of radiation, the calculation method of the reflectivity [5–8] is based on the solution of the Helmholtz equation for the complex amplitude of the electric field $E(z)$ with the wavelength $\lambda$, which propagates in the medium with nonuniform $\varepsilon(\lambda, z)$

$$\frac{d^2E(z)}{dz^2} + \frac{4\pi^2}{\lambda^2} \varepsilon(\lambda, z)E(z) = 0$$

In the equation (8) $\lambda$ is the wavelength of the electromagnetic wave in vacuum and the amplitude $E(z)$ is dimensionless function. The equation (8) is solved in the region $z \in [0, h]$ with the corresponding boundary conditions

$$E(0) = \exp \left(2\pi i \sqrt{\varepsilon(0)} \frac{h}{\lambda}\right)$$

$$E'(0) = (2\pi i / \lambda) \sqrt{\varepsilon(0, \lambda)} E(0),$$

where $h$ is the width of the optically non-uniform region. Schematic density profile and the spatial dependence of $\varepsilon(\lambda, z)$ are shown in figure 2. Region I ($z < 0$) is vacuum, where $\varepsilon(\lambda, z) = 1$. Optically non-uniform medium $z \in [0, h]$ is region II in figure 2. Region III corresponds to considered medium with uniform DF. Since the equation (8) is solved in region II, the linear approximation for the DF is used $\varepsilon(\lambda) = 1 + (\varepsilon(\lambda, h) - 1) \cdot (z/h)$. The value of the $\varepsilon(\lambda, h)$ is calculated within the framework of the DFT by formulas (6), (7). In this case the Fresnel formula (1) is inapplicable and reflectivity is found from the following expression

$$R = \left|\frac{(2\pi i E(0) - \lambda E'(0))}{(2\pi i E(0) + \lambda E'(0))}\right|^2.$$
Figure 2. Schematic density profile and corresponding spatial dependence of the DF $\varepsilon_\lambda(z)$. Region I is vacuum. Region II corresponds to optically non-uniform medium (broadened shock wave front) with finite width $h$. Region III is considered medium (plasma).

Figure 3. Dependencies of the shocked xenon plasma reflectivity on the plasma density at various values of wavelengths 694 nm (a) and 532 nm (b). The experimental data [2, 8] are depicted by stars. The squares correspond to the calculation data. The arrows indicate the values of the plasma density where the frequency of incident radiation and the plasma frequency coincide with each other. The dashed lines are calculated reflectivities from the broadened wave front.

The measured and calculated values of the reflectivity dependence on density are shown in figure 3 for the wavelengths of laser radiation 694 nm (a) and 532 nm (b). The experimental data [2, 8] are depicted by stars. The squares correspond to calculation results obtained in [9, 27] without introduction of the wave front broadening. As one can see, the calculated reflectivities for the wavelength 694 nm (figure 3a) are in a good agreement with the experimental data both in the absolute values and in the density dependence. Theoretical values of the reflectivity at 532 nm are overestimated in comparison with the experimental ones. However, the relative dependence of the reflection coefficient on the density is reproduced.

The wave front width $h$ is parameter of the equation (8). It is possible to choose the range of the values of $h$, which gives the exact experimental values of reflectances at each density within the error bars (the range of $h$ is determined by the experimental error of measurement of reflectivity.). The values of the width $h$ are given in table 1. In figure 3 the dependence of reflectivity on density calculated by formula (10) is depicted by dashed line. The introduction of the wave front broadening improves the agreement with the experiment for wavelength $\lambda=532$ nm and at the same time it does not make worse the agreement for $\lambda = 694$ nm. Therefore, since
Table 1. The width of the wave front \( h \) (nm) for various values of the density \( \rho \) (g/cm\(^3\)).

| \( \rho \) | 1.1 | 1.6 | 2.2 | 2.8 | 3.4 |
|---|---|---|---|---|---|
| \( \lambda = 694 \text{ nm} \) | 0–135 | 60–145 | 40–112 | 0–110 | 0–25 |
| \( \lambda = 532 \text{ nm} \) | 130–180 | 130–175 | 96–120 | 75–115 | — |

the values of the width are close to the theoretical value 100 nm, which is physically justified, the approach gives complete explanation of the experimental data [2, 8].

3.2. Polarized reflectivity

For calculation of dependence of the s-polarized electric field \( E(z) \) amplitude on incident angle \( \varphi \) the expression \( \varepsilon(\lambda, z) - \sin^2 \varphi \) has to be substituted in (8) and in boundary conditions (9) instead of \( \varepsilon(\lambda, z) \). The amplitude of p-polarized magnetic field \( H(z) \) is determined by the following equation

\[
\varepsilon(\lambda, z) \frac{\partial}{\partial z} \left( \frac{1}{\varepsilon(\lambda, z)} \frac{\partial H(z)}{\partial z} \right) + (\varepsilon(\lambda, z) - \sin^2 \varphi) H(z) = 0
\]

Substituting in (11) instead of \( \varepsilon(\lambda, z) \) the p-polarized reflectivity is found from the following expression

\[
R = \left| \frac{2\pi i \sqrt{1 - \sin^2 \varphi} H(0) - \lambda H'(0)}{2\pi i \sqrt{1 - \sin^2 \varphi} H(0) + \lambda H'(0)} \right|^2
\]

For calculation of the s-polarized reflectivity the corresponding values of the amplitude \( E(0) \) and its derivative \( E'(0) \) have to be substituted in (12).

The depth of the wave front \( h \) can be also estimated within the framework of the Drude theory [31–33] for polarized reflection of the electromagnetic wave using the experimental dependence of \( R_s \) and \( R_p \) on incident angle \( \varphi \). For this approach the magnitude of \( h \) can be evaluated from equation

\[
\frac{R_p}{R_s} = \pi^2 \cdot \frac{h^2}{\lambda^2} \left| \frac{n^2 + 1}{(n^2 - 1)^2} \beta^2 \right|
\]

where \( \beta = \sqrt{N^2 + n^2 \left( \frac{1}{N^2} \right)} 
- 1 - n^2 \), \( N = N(z) = \sqrt{\varepsilon(z)} \) is the nonuniform refraction coefficient, symbol is averaging over the width of the transitive layer (wave front), \( n \) is refraction coefficient of the plasma. The ratio \( R_s/R_p \) is found at value of angle \( \varphi \) where \( R_p \) reaches the minimum (Brewster angle). For linear dependence of \( N(z) \) the expression (13) gives the following result for \( h \)

\[
h = \lambda \left( \frac{3}{2\pi} \right) \left( \frac{R_p}{R_s} \right)^{1/2} (R \cdot |n^2 + 1|)^{-1/2}
\]

where \( R \) is reflection coefficient for the normal incidence. The refraction coefficient \( n = \sqrt{\varepsilon} \) is calculated within the framework of the DFT.

The measured and calculated values of the \( R_s \) and \( R_p \) reflectivity dependence on incident angle \( \varphi \) are shown in figure 4 for the wavelengths of laser radiation 1064, 694 and 532 nm and densities 2.7 and 2.8 g/cm\(^3\). The experimental data [3, 4] are depicted by squares \( (R_s) \) and circles \( (R_p) \). The solid lines correspond to the calculated results obtained in this work within framework of the DFT with the longitudinal expression (6) for the imaginary DF and without introduction of the broadening of the wave front. The calculated minimum of the dependence
The self-consistent approach for optical and electronic properties of warm dense matter and nonideal plasmas [9–13] is extended for description of reflectance from the broadened shock wave front.

Within the framework of the approach, the method of calculation of reflectivity, which is based on solution of the Helmholtz equation for the amplitudes of the electromagnetic field, is applied. The method allows taking into account non-uniformity of the density profile (shock wave front broadening) and is used for calculation of normal reflectivity as well as for s- and p-polarized reflectivities.

The method based on Drude theory [31,32] of reflection is considered. It allows calculating the ratio of the s- and p-polarized reflectances, if dependence of the dielectric function on distance is known.

The reflectivities calculated with taking into account of the shock wave broadening are in a good agreement with the experimental data for xenon plasmas [1–4]. The inverse problem is solved: the width of the shock wave front is estimated based on calculated values of reflectivity and dielectric function. The width of the shock wave front is estimated as a parameter of solution of the Helmholtz equation. The additional method of estimation of the width is suggested. It allows estimating wave front width from experimental ratio of s- and p-polarized reflectivities within the framework of Drude theory of refraction from optically nonuniform medium. Both values of the wave front width are closer to the physically approved width of propagation of electron avalanche in shock compressed xenon contrary to the results [4,7,8].

Figure 4. Dependencies of the shocked xenon plasma $R_s$ (green) and $R_p$ (red) reflectivities on the incident angle at various values of wavelengths and density. The experimental data [3, 4] are depicted by squares ($R_s$) and circles ($R_p$). The solid lines correspond to the calculation results, which are obtained without introduction of the wavefront broadening. The dashed lines correspond to the calculated values of the reflectivity with the broadened wavefront.

$R_p(\phi)$ is shifted relatively the experimental one. It can be considered as notification of existence of the transitive region with finite width, where the plasma density increases smoothly to a final value. Dashed lines correspond to the results calculated with introduction of the wave front broadening.

The method of estimation of the shock front width described in the previous subsection gives the magnitude of $h = 95$ nm at $\rho = 2.8 \, \text{g/cm}^3$. The results calculated with introduction of this wave front broadening shown by dashed lines in figure 4. The widths calculated by formula (14) are approximately 150 nm. As one can see this method also gives estimation of the width, which are less than estimations of [3, 4] and closer to theoretical value 100 nm.

4. Conclusions
The self-consistent approach for optical and electronic properties of warm dense matter and nonideal plasmas [9–13] is extended for description of reflectance from the broadened shock wave front.
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