Hydrodynamic Simulation of the Femtosecond Laser Processing of Metals

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ABSTRACT

The electron–ion collision frequency is a key factor for simulations of the interaction process of ultrashort lasers with metals. In this paper, we propose a semiempirical method based on the Drude–Sommerfeld model for obtaining the electron–ion collision frequency over a wide temperature range, from solid-state to high-temperature plasma. Moreover, we performed a series of hydrodynamic simulations including the modified electron–ion collision frequency. The results indicated that the modified electron–ion collision frequency can exactly describe not only the energy deposits of various femtosecond laser pulses but also the transient properties of electrons on the metal surface. Moreover, a simple function was identified to describe the relationship between laser intensity and peak electron temperature on the surface. This method is expected to be beneficial for simulations of the interaction process of femtosecond laser pulses with metals.

INTRODUCTION

During the past two decades, ultrashort laser heating of metals has been a very active topic for research due to its large number of applications. (1-8) The ultrahigh instantaneous power and ultrashort pulse duration of femtosecond laser pulses produces nonlinear(9, 10) and nonequilibrium processes. (11, 12) Several models based on different theories have been developed to study the interaction processes and subsequent dynamic evolution of the target. Molecular dynamics (MD) is a suitable tool for studying phase change and material removal. (13-15) As the fundamental input in MD simulations, the potential function must be obtained through the quantum mechanics method. The particle-in-cell method solves the equations of motion and Maxwell’s equations for particles in order to determine the forces among these particles. (16-18) which is very similar to the description of plasma from first principles as a system of charged particles. However, this method can be computationally expensive. A hydrodynamic (HD) model has been developed to describe the expansion and heating of targets irradiated by laser pulses. (19, 20) It achieves a balance between the calculation of material properties and the time required.

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The HD motion coupled with laser propagation and several transport mechanisms are solved using one-dimensional geometry and a fractional step scheme. Fluid motion, as well as heat diffusion, is solved using the implicit Lagrangian method. Transport by thermal conduction and radiation, as well as electron–ion energy transfer, is described in the two-temperature (electron and ion) model,(21) which covers a wide range of laser intensities from solid-state to high-temperature plasma. Laser propagation is calculated using the one-dimensional Maxwell’s equations, and radiation transfer is solved using the forward-reverse method for a discrete number of frequency groups. Matter properties are interpolated from tables (equations of state, ionization, opacities, and emissivities) that are generated using external codes.(22-25)

Electron–ion collision frequency is the most important physical quantity in the context of HD simulations; it governs laser energy absorption, electron–ion energy transfer, electronic heat conduction, electrical conductivity, and the energy loss of ion beams. In this paper, a semiempirical method based on the Drude–Sommerfeld model(26) is proposed to obtain precise electron–ion collision frequency measurements in a wide range of temperatures, from solid-state to high-temperature plasma. Meanwhile, the modified electron–ion collision frequency is employed to describe the transient properties of electrons on the metal surface irradiated by various femtosecond laser pulses.

METHODS

The detailed description of the theory and its implementation can be found in Ref (27). The theory is described briefly as follows.

Laser energy deposition

In contrast to nanosecond laser pulses, femtosecond laser pulses generate steep gradient plasmas with typical scale lengths in the order of the laser wavelength. Under these conditions, Maxwell’s equations can be solved explicitly to obtain the light field. In Gaussian units, Maxwell’s equations have the following form:

\[ \nabla \times \mathbf{E} + \frac{1}{c} \frac{\partial \mathbf{B}}{\partial t} = 0, \quad \nabla \cdot \mathbf{E} = 4\pi \rho \]

\[ \nabla \times \mathbf{B} - \frac{1}{c} \frac{\partial \mathbf{E}}{\partial t} = \frac{4\pi}{c} \mathbf{j}, \quad \nabla \cdot \mathbf{B} = 0 \]

(1)

Furthermore, assuming that the current density is linearly dependent on the electric field, we can decompose any generic field quantity \( \mathbf{A} \) into the sum of periodic terms. For monochromatic irradiation (\( \mathbf{A} = \Re(\hat{\mathbf{A}} e^{-i\omega t}) \)), Eq. (1) is reduced to

\[ \nabla \times \hat{\mathbf{E}} + i\frac{\omega}{c} \hat{\mathbf{B}} = 0, \]

\[ \nabla \times \hat{\mathbf{B}} - i\frac{\omega}{c} \hat{\mathbf{E}} = \frac{4\pi}{c} \hat{\mathbf{j}}. \]

(2)

The relationship between the current density \( \hat{\mathbf{j}} \) and the electric field is given by Ohm’s law, \( \hat{\mathbf{j}} = \sigma_r \hat{\mathbf{E}} \). Then, Maxwell’s equations are written as follows:
\[ \nabla \times \hat{E} + \frac{i\omega}{c} \hat{B} = 0, \]
\[ \nabla \times \hat{B} - \frac{i\omega \sigma}{c} \hat{E} = 0. \]

where \( \varepsilon_r = 1 + \frac{4\pi i \sigma_r}{\omega} = 1 - \frac{\omega_p^2}{\omega^2(1 + i\nu_c/\omega)} \) is the transversal dielectric function and \( \omega_p = (4\pi e^2 n_e/m_e)^{1/2} \) is the plasma frequency. The mean energy flux in the \( z \) direction is obtained by averaging the \( z \) component of the Poynting vector \( \frac{c}{4\pi} \mathbf{E} \times \mathbf{B} \) over one laser oscillation period, \( T = 2\pi/\omega \). Thus, the mean energy flux is written as follows:

\[ \phi = \frac{1}{T} \int_0^T \left( \frac{c}{4\pi} \mathbf{E} \times \mathbf{B} \right) dt \]

The specific power deposition at cell centers is computed from the difference of fluxes as follows:

\[ \left( \frac{D}{\rho} \right)_j = \frac{\phi_j - \phi_{j+1}}{m_j} \]

Hydrodynamics

The fluid description is based on conservation laws for mass, momentum, and energy:

\[ \frac{\partial \rho}{\partial t} + \nabla \cdot \mathbf{v} = 0, \]
\[ \rho \frac{\partial \mathbf{v}}{\partial t} = \nabla \cdot \mathbf{\sigma}, \]
\[ \rho \frac{\partial e}{\partial t} = \mathbf{\sigma} : \nabla \mathbf{v} - \nabla \cdot \mathbf{S}_{th} + D_{ext}. \]

In these equations, the operator \( \partial / \partial t \equiv \partial / \partial t + \mathbf{v} \cdot \nabla \) denotes the time derivation in a frame that is fixed in relation to the fluid; \( \rho, \mathbf{v}, \, e, \mathbf{\sigma}, \) and \( \mathbf{S}_{th}, \) are the mass density, fluid velocity, specific internal energy, stress tensor, and thermal flux, respectively. The source term \( D_{ext} \) represents energy deposition by the laser pulse.

The stress tensor is the sum of the thermodynamic equilibrium pressure and a so-called artificial viscosity that describes the shock waves.

\[ \mathbf{\sigma} = -(P(\rho, e) + P_v) \mathbf{U} \]

Here, \( \mathbf{U} \) is the unit second rank tensor, and

\[ P_v = \begin{cases} 2\rho L^2 (\nabla \cdot \mathbf{v})^2, & \nabla \cdot \mathbf{v} < 0 \\ 0, & \nabla \cdot \mathbf{v} \geq 0 \end{cases} \]

where \( L \) is a characteristic length equal to the cell thickness. The equations of state \( (P = P(\rho, e), T = T(\rho, e)) \) are generated using external codes. (22-25) For the plasmas considered in this study, the energy deposition from the laser pulse induces a
nonequilibrium between electrons and ions. Therefore, the additive splitting of Eq. (8) into an electron and an ion equation of state is valid; Eq. (6)–(8) can be rewritten as follows:

\[
\frac{D \rho}{Dt} = -\rho \nabla \cdot \mathbf{v} \\
\rho \frac{D \mathbf{v}}{Dt} = -\nabla (P_e + P_i + P_i) \\
\rho \frac{D e_e}{Dt} = -P_e \nabla \cdot \mathbf{v} - \nabla \cdot \mathbf{S}_{th} + Q_{ei} + D_{ext} \\
\rho \frac{D e_i}{Dt} = -(P_i + P_i) \nabla \cdot \mathbf{v} - Q_{ei}
\]

Where the relaxation term \( Q_{ei} \) is proportional to the temperature difference between electrons and ions.

**Thermal conductivity**

In plasma with gentle temperature gradients, the electron thermal flux is given by

\[
\mathbf{S}_{th} = -K \nabla T_e. \tag{15}
\]

The thermal conductivity is determined by the electron–ion collision frequency:(25)

\[
K = \alpha(Z) \frac{n_e k^2 T_e}{m_e v_e}, \tag{16}
\]

where \( n_e = Z p / (A^m m_p) \) is the electron number density; \( Z \) and \( A^m \) are the averaged ion charge and mass number, respectively; \( k \) is the Boltzmann constant; \( T_e \) is the electron temperature; \( m_e \) and \( m_p \) are the masses of the electron and proton, respectively; and \( v_e \) is the electron collision frequency. The numerical coefficient \( \alpha(Z) \) is obtained from the ion number \( Z \) as follows:

\[
\alpha'(Z) = 3.22554 \frac{Z + 0.24}{1 + 0.24 Z}. \tag{17}
\]

Under short pulse laser irradiation, extreme temperature gradients may occur, and the heat flux is limited according to the following inequality(28):

\[
|S_{th}| \leq S_{th}^{max} = f n_e k T_e \sqrt{\frac{k T_e}{m_e}}, \tag{18}
\]

where the flux limit \( S_{th}^{max} \) is set to the physical maximum reduced by an empirical factor \( f \). (29) This limitation is implemented by replacing \( K \) with the following modified conductivity:

\[
K' = \frac{K S_{th}^{max}}{S_{th}^{max} + K |\nabla T|}, \tag{19}
\]

**Electron–ion collision frequency**

The electron–ion collision frequency determines the laser energy absorption, electron–ion energy transfer, electronic heat conduction, and electrical conductivity. For a cold solid, the collision frequency is determined by the electron–phonon collision frequency. For the ideal plasma, it is obtained using a well-established classical theory.
However, for warm dense matter (WDM) and metal-like solids, it is difficult to obtain the
electron–ion collision frequency when electrons degenerate and other quantum behavior,
involving bound electrons and strong ion–ion correlations, is introduced.

An approach based on the Drude–Sommerfeld model that attributes the dominant part
of the collision frequency to electron–ion collisions and considers the electron degeneracy
in WDM and the modifications due to Coulomb scattering can accurately describe the
dynamic collision frequency. In the WDM regime, the collision frequency is given as follows:

\[
\nu_{\text{WDM}} = \frac{4\pi^2 \Gamma^2(1/3)}{15 \cdot 3^5 \Gamma(2/3)} \frac{Z n_e}{m_e^2 v_{th}^3} \left( \frac{m_v v_{th}}{Ze^2 \omega} \right)^{2/3} \left( \frac{v_{th}}{v} \right)
\]

where \( \Gamma(x) \equiv \sqrt{1 - x (16 + 8x + 6x^2)} / 15 \), \( Z \) is the ion charge, \( n_e \) is
the electron number density, \( m_e \) is the electron mass, \( v_{th} = (kT_e / m_e)^{0.5} \) is the electron
thermal velocity, \( \omega = \max(\omega_e, \omega_p) \), and \( \omega_p = (4\pi n_e e^2 / m_e)^{0.4} \) is the plasma frequency.

Moreover, \( \left( \frac{v_{th}}{v} \right) \) represents Pauling blocking and is given as follows:

\[
\frac{v_{th}}{v} = \frac{3v_{th}}{m v_F^3} \int_0^\infty f(\varepsilon) \left( 1 - f(\varepsilon + h\omega) \right) d\varepsilon
\]

\[
= 3 \left( \frac{v_{th}}{v_F} \right)^3 \ln \left( \frac{1 + e^y}{1 + e^{y-z}} \right) \left( \frac{1}{1 - e^{-z}} \right)
\]

where \( y = \mu / kT_e \), \( z = h\omega / kT_e \), and \( v_F = (3\pi^2 n_e)^{1/3} h / m_e \) is the characteristic Fermi
velocity of the degenerate electron gas.

The overall collision frequency is obtained by interpolating between the WDM and
ideal plasma regimes as follows:

\[
\nu_e(T_e, \omega) = 2\sqrt{2\pi} \frac{Ze^4 n_e}{m_e^{1/2} (kT_e)^{3/2}} \times \ln \left[ 1 + K_{ds} \frac{1.32}{\sqrt{2\pi} \left( m_e^{1/2} Ze^2 \omega \right)^{2/3}} \right] F(T_e, h\omega)
\]

where, the Fermi factor \( F(T_e, h\omega) = \sqrt{\pi / 2} \left( v_{th} / v \right) \) assumes the following asymptotic forms:

\[
F(T_e, h\omega) \rightarrow \begin{cases} 
3 \left( \frac{v_{th}}{2} \right) \min \left( \frac{h\omega}{kT_F}, 1 \right) & \text{for } T_e \rightarrow 0 \\
1 & \text{for } T_e \rightarrow \infty 
\end{cases}
\]

The parameter \( K_{ds} \) may account for solid-state effects, such as an effective electron
mass, and it can be used to adjust the model according to measured values. We propose a
semiempirical method through which the collision frequency can be modified with
adjustable parameters:

\[
v_m = \begin{cases} 
\nu_e & \text{for } Z < Z_{critical} \\
\exp \left[ \frac{\nu_e A}{B - 2T_e / T_F} \right] + 1 & \text{for } Z \geq Z_{critical}
\end{cases}
\]
RESULTS AND DISCUSSION

A HD simulation was performed using the MULTI-fs code,(27) a numerical tool for studying the interaction of ultrashort subpicosecond laser pulses with matter in the intensity range from $10^{11}$ to $10^{17}$ W/cm$^2$. A schematic of the model setup is shown in Fig. 1. The metal film was located in the region given by $x > 0$, and the upper surface was aligned with $x = 0$. The laser beam traveled in the $x$ direction, and it irradiated perpendicular to the target.

Figure 1. Schematic of the model setup.

To obtain the modified electron–ion collision frequency, we performed a set of HD simulations for three metals (Al, Au, and Cu) with a thickness of 550 nm that were irradiated by femtosecond laser pulses with various intensities.(30) Figs. 2(a)–(c) show the absorption ratios of Al, Cu, and Au, respectively, at different laser intensities. The Drude–Sommerfeld model provides precise absorption ratios only in the high-intensity region. Laser energy absorption, which is determined from the imaginary part of the complex dielectric constant given in Eq. (3), is an extremely complex process, especially for femtosecond laser pulses. To describe the energy absorption, the collision frequency must be determined over a broad temperature range. For the high-intensity region, in which the electron temperature is substantially higher than the Femi temperature $T_F$, the energy absorption was defined according to the Coulomb electron–ion collision frequency, which the Drude–Sommerfeld model describes accurately. However, for the mild intensity range, the parametric dependence of collision frequency $v_e$ on temperature changed completely. Herein, we propose a semiempirical method based on the Drude–Sommerfeld model. By selecting suitable parameters, we can obtain the modified electron–ion collision frequency, which accurately describes laser energy absorption in the wild intensity range. The dependence of parameters on material is shown in Table 1.

Table 1. Parameters for the three metals.

<table>
<thead>
<tr>
<th>metal</th>
<th>$Z_{\text{critical}}$</th>
<th>A</th>
<th>B</th>
<th>C</th>
</tr>
</thead>
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<tr>
<td>Al</td>
<td>0.3</td>
<td>1.2</td>
<td>1.5</td>
<td>1</td>
</tr>
<tr>
<td>Cu</td>
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<td>8</td>
<td>0.2</td>
<td>1</td>
</tr>
<tr>
<td>Au</td>
<td>0.35</td>
<td>4</td>
<td>0.4</td>
<td>2</td>
</tr>
</tbody>
</table>
Figure 2. Absorption rate of (a) Al, (b) Cu, (c) and Au based on a modified Drude–Sommerfeld model (red solid line), a Drude–Sommerfeld model (black dash line), and experiments (30) (black solid line).
To illustrate the modified electron–ion collision frequency, we conducted a set of HD simulations for different metals irradiated by various femtosecond laser pulses. We considered a copper film with a thickness of 550 nm irradiated by a femtosecond laser pulse with a duration of 50 fs, wavelength of 800 nm, and various intensities ($1 \times 10^{13}$, $1 \times 10^{14}$, and $1 \times 10^{15}$ W/cm$^2$). Electron temperature at the target surface as a function of time is shown by the dashed lines in Fig. 3, whereas the results of another study(31) are represented by the solid lines. The two results were correlated closely at different intensities, indicating that the modified electron–ion collision frequency can accurately describe the transient properties of electrons on the copper surface. Additionally, Fig. 3 indicates that the electron system heated rapidly after the laser pulse reached the target surface, and the electron temperature increased to the peak value after approximately 80 fs. The dependence of the highest electron temperature on the laser intensity can be determined using $T_e = kI^{3/5}$, where $T_e$ is the electron temperature, $I$ is the laser intensity, and $k = 3.17 \times 10^{-8}$. Figure 4 shows the electron temperature profile versus time for the intensity of $1 \times 10^{13}$ W/cm$^2$; the black line represents the normalized intensity envelope of the applied femtosecond laser pulse. As indicated in the figure, the laser energy was initially deposited into electrons at the surface layer, after which the inner electrons were heated by the electron–electron collision. However, after 100 fs, the electron temperature decreased because of the electronic thermal diffusion effect.
The dependence of the energy deposition of femtosecond laser pulses on the pulse duration was also studied. Figure 5 shows the absorption ratio of an Al film for various laser intensities and different durations. The peak absorption ratio was reduced slightly and moved toward the mild intensity region as the pulse duration increased. However, the influence of pulse duration became trivial when fluence was considered (Fig. 6). This was because the duration of the femtosecond laser pulse was substantially shorter than the electron–ion relaxation time. The laser energy resided predominantly in the thermal energy of electrons. The electron–ion collision frequency has the same parametric dependence on temperature and other parameters, even for laser pulses with different durations when the target is irradiated by laser pulses with equal energy. Therefore, pulse duration has little effect on the energy absorption for a femtosecond pulse.\(^{(32)}\)
Figure 7. Spatial dependences of electron temperature ($T_e$), ion temperature ($T_i$), mass density ($\rho$), pressure ($p \equiv P_e + P_i$), free electron density ($n_e$), and laser energy deposition ($De/v$) at 150 fs.

Figure 8. Motion of Lagrangian interfaces as a function of time.

Figure 7 displays the spatial profiles of various variables of the Al film for an intensity of $10^{15}$ W/cm$^2$. Figure 8 shows the motion of the Lagrangian interface. The mesh had finer zoning toward the irradiated side where the interaction occurred. The absorbed energy (approximately 22% of the laser light) resided predominantly in the thermal energy of electrons, which were heated to almost 30 eV. Additionally, a small fraction of the thermal energy was translated into kinetic energy, which caused the front layers to expand rapidly, with a velocity of up to $7.8 \times 10^3$ m/s (Fig.8). The ion temperature was lower than the electron temperature by a factor of 15 because of the slow electron–ion relaxation. However, ions were heated to equilibrium through the electron–ion energy transfer.(8) The peak pressure reached almost 20 Mbar at 3 nm, where the maximum specific laser energy deposition occurred. The free electron density increased rapidly with the solid density and reached a peak value five times higher than that at room temperature.

CONCLUSIONS

We propose a semiempirical method based on the Drude–Sommerfeld model; in this method, the collision frequency between electrons and ions can be modified for a wide range of electron temperatures from solid-state to high-temperature plasma. Additionally, a series of HD simulations illustrated that the modified electron–ion collision frequency can exactly describe the transient properties of electrons on the metal surface and the thermodynamic process of the metal film. This method is expected to be beneficial for the simulations of interaction processes between femtosecond laser pulses and metals.
simple function was identified to describe the relationship between laser intensity and peak electron temperature on the surface.

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