Ultrashort laser pulse energy deposition in metal films with phase changes

Yunpeng Ren, J. K. Chen,a) Yuwen Zhang, and Jing Huang
Department of Mechanical and Aerospace Engineering, University of Missouri, Columbia, Missouri 65211, USA

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Four optical models of reflectivity and absorption coefficient are investigated in this letter. After
compared with existing experimental data, the extended Drude model is incorporated into a
two-temperature model to simulate laser energy deposition and thermal response, including solid–
liquid and liquid–vapor phase change, in a gold film irradiated by a femtosecond laser pulse.
Dynamic reflectivity and absorption coefficient should be employed in modeling ultrafast laser
heating except for very low laser fluencies. © 2011 American Institute of Physics.

Rapid advancement in ultrashort-pulsed lasers over the
last two decades has been driving a new wave of research in
many areas in physics, chemistry, bioscience, and
materials.1–5 To date, temperature-dependent thermophysical
properties have been employed in simulating ultrafast laser
heating.6,7 However, the optical properties receive much less
attention. Recently, the transient nonlinear absorption in Au
films has been found to be directly related with the thermal
smearing of the electron distribution by Rotenberg et al.8
using the pump-probe experiment. In this letter, we investi-
gate temperature dependences of reflectivity (R) and absorption
coefficient (α) and their impacts on prediction of ul-
trafast thermal response in metal films, including solid–
liquid (S–L) and liquid–vapor (L–V) phase change.

Drude model is generally expressed as follows:9

\[
\varepsilon_D = \varepsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma_p)} = \varepsilon_1(x,t) + i\varepsilon_2(x,t),
\]

where \(\varepsilon_\infty\) is a dc dielectric function, \(\omega\) plasma frequency, \(\omega\) laser frequency, and \(\gamma_p\) damping coefficient that is equal to reciprocal of electron relaxation time (\(\tau_e\)). The above model is referred to simple Drude model if \(\varepsilon_\infty = 1\), otherwise modified Drude model. The reflectivity and absorption coefficient can be determined in terms of \(\varepsilon_1\) and \(\varepsilon_2\).

Recently, Vial et al.10 pointed out that the Lorentz–
Drude model11 with five Lorentzian terms12 cannot match
well experimental data around 2 eV for gold (the energy for interband transition from d-band to Fermi level in gold is \(E_F - E_d = 2.38\) eV). They thus proposed an extended Drude model as follows:

\[
\varepsilon_{DL} = \varepsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma_p)} - \frac{f\Omega_L^2}{(\omega^2 - \Omega_L^2 + i\Gamma_L\omega)},
\]

where \(\Omega_L\) and \(\Gamma_L\) stand for the oscillator strength and the spectral width of the Lorentz oscillators, respectively, and \(f\) is a weighting factor. With the values optimized for gold; \(\varepsilon_\infty = 5.9673\), \(\omega_p = 1.328 \times 10^{16}\) Hz, \(\Omega_L = 4.085 \times 10^{15}\) Hz, \(\Gamma_L = 0.659 \times 10^{15}\) Hz, and \(f = 1.09\), their model fitted experimental data quite well in the range of 1.24–2.48 eV.

In modeling ultrashort pulse laser interaction with met-
als, Eesley14 and recently Jiang and Tsai15 added an inter-
band transition part to the imaginary part of electrical permittivity in the simple Drude model.

Figures 1 and 2 compare \(R\) and \(\alpha\) for gold at room tem-
perature (RT), respectively, among four optical models. The
electron relaxation time is calculated by16

\[
\tau_e = \frac{1}{A_eT_e^2 + B_eT_l},
\]

where \(T_e\) and \(T_l\) are electron and lattice temperature, respec-
tively, and \(A_e = 1.2 \times 10^7 \text{ K}^{-2} \text{s}^{-1}\) and \(B_e = 1.23 \times 10^{11} \text{ K}^{-1} \text{s}^{-1}\) for gold. For 250–1000 nm, the Lorentz–
Drude model13 fits fairly well with the experimental data.17
On the other hand, the extended Drude model matches very
well for 500–1000 nm. This is because the parameter opti-

ization depends on the smoothness of data over the energy
range of interest. Since the laser pulse of interest here is in
the range of 500–1000 nm, the extended Drude model is
considered in the following analysis.

The reflectivity would decrease significantly when \(\tau_e < 5\) fs, and \(\alpha\) when \(\tau_e < 2\) fs. The significant changes in \(R\) and \(\alpha\) could drastically alter laser energy deposition and in turn, thermal response in a target. The \(\tau_e\) corresponding to \(\tau_e = 5\) fs is 3.617 \times 10^3 K based on \(T_l = 350\) K. This temperature state can be reached in a gold film heated by a 100 fs laser pulse at a low fluence 0.075 J/cm².

FIG. 1. (Color online) Reflectivity vs wavelength at RT calculated by dif-
erent optical models.
To study the impacts of $R$ and $\alpha$ on thermal response of metals under ultrafast laser heating, we consider a 1 $\mu$m gold film irradiated on the front side ($x=0$) by a 100 fs, 600 nm laser pulse of 0.2 J/cm$^2$. The initial temperature is set at 300 K.

The energy equations of a two-temperature model\textsuperscript{7,8} are given by

\begin{equation}
C_p \frac{\partial T_e}{\partial t} = \frac{\partial}{\partial x} \left( k_e \frac{\partial T_e}{\partial x} \right) - G(T_e - T_l) + S(x,t),
\end{equation}

\begin{equation}
C_l \frac{\partial T_l}{\partial t} = \frac{\partial}{\partial x} \left( k_l \frac{\partial T_l}{\partial x} \right) + G(T_e - T_l),
\end{equation}

where the subscripts $e$ and $l$ are associated with electrons and lattice, respectively. The volumetric laser heat source is described by the following equation:\textsuperscript{15}

\begin{equation}
S(x,t) = 0.94 J_e \frac{1 - R(0,t)}{t_p} \alpha(x,t) \exp \left[ - \int_0^x \alpha(x,t) dx \right] - 2.77 \left( \frac{t}{t_p} \right)^2,
\end{equation}

where $J_e$ and $t_p$ are laser pulse fluence and pulse duration, respectively.

The energy balance at the S–L interface is described by\textsuperscript{19}

\begin{equation}
k_s \frac{\partial T_{s,l}}{\partial x} - k_l \frac{\partial T_{l,l}}{\partial x} = \rho_l h_v u_{s,l} dT_{s,l},
\end{equation}

where the subscripts $s$ and $l$ are associated with solid and liquid, respectively; $\rho$ is mass density; $h_v$ is latent heat of fusion; $u_{s,l}$ is the S–L interfacial velocity. For a rapid melting/solidification process, the S–L interfacial velocity is controlled by nucleation dynamics described by\textsuperscript{20}

\begin{equation}
u_{s,l} = u_0 \left[ 1 - \exp \left( - \frac{h_v m}{R c_{\text{v,liquid}}} \frac{T_{s,l} - T_m}{T_{s,l}} \right) \right],
\end{equation}

where $u_0=1300$ m/s is the maximum interfacial velocity, $R$ is the gas constant for metals, and $T_{s,l}$ is the interfacial temperature.

For the L–V interface, the energy balance equation at the L–V interface is

\begin{equation}\rho_l h_v u_{e,l} + \sigma \varepsilon (T_{e}^4 - T_{l}^4) = - k_l \frac{\partial T_l}{\partial x},
\end{equation}

where $h_v$ is latent heat of evaporation; $u_{e,l}$ and $T_{e,l}$ are L–V interfacial velocity and temperature, respectively.

The vaporization wave model,\textsuperscript{21} proposed for superheating kinetics of volume vaporization in electrically exploded wires, is employed to determine the L–V interfacial velocity. According to shock wave theory,\textsuperscript{22} the velocity of dynamic evaporation is limited by the characteristic speed of sound at the liquidus line given by\textsuperscript{23}

\begin{equation}
\nu_s = \frac{dp}{dT}\left( \frac{T}{\rho c_{\text{v,liquid}}} \right)^{\frac{3}{2}},
\end{equation}

where $\rho_s$ and $c_{\text{v,liquid}}$ are density and specific heat of liquid, respectively. The determination of $dp/dT$ can be found in Ref. 24.

Together with the adiabatic boundary conditions except for radiation heat loss at the irradiated surface, the above equations are solved using a finite difference method. The detailed solution procedure can be found in Huang et al.\textsuperscript{7}

The electron–phonon coupling factor employed here is from Lin and Zhigilei\textsuperscript{2} and all other temperature-dependent material data are from Huang et al.\textsuperscript{7}

Figure 3 shows the changes in $R$ and $\alpha$ at $x=0$ during the lasing. The normalized temporal profile of the laser pulse and the normalized $\tau_e$ are also presented. The reflectivity starts to decrease considerably after $t=60$ fs, while $\alpha$ after $t=40$ fs. Both the minimum $R$ and $\alpha$ are present at $t=95$ fs, the time when the peak $T_e$ occurs. After that, $\tau_e$ increases gradually due to the combined effect of decreasing $T_e$ and increasing $T_p$. As a result, both $R$ and $\alpha$ keeps on a small increase until the laser is off.

The distributions of laser heat source ($S$) at different times are shown in Fig. 4. The absorbed laser energy density at $x=0$ reaches its peak value at $t=-16.5$ fs, about 5.2 times the peak value $0.88 \times 10^{17}$ W/cm$^2$ computed with $R=0.93$ and $\alpha=67.89$ $\mu$m$^{-1}$ at RT. The volumetric laser heat distributions after $t=-16.5$ fs differ significantly from Beer’s law.

Figure 5 presents the electron and lattice temperature as functions of time. Obviously, both $T_e$ and $T_p$ simulated with the dynamic $R$ and $\alpha$ are much higher than their counterparts obtained with the constant values at RT. The peak $T_e$ and $T_p$
are 33 572 K and 4278 K versus 13 196 K and 857 K, respectively. For the latter, the underestimated \( T_e \) is well below the melting point. The result that \( T_e \) is lower than \( T_L \) after about \( t=12 \) ps is because thermal energy in electrons diffuses away much faster than that in the lattice. In summary, use of accurate optical properties is crucial in modeling high-power ultrashort laser interaction with matter. In this letter, we compare four optical property models and then use the extended Drude model to calculate \( R \) and \( \alpha \). The numerical analysis is performed for a thin gold film and then use the extended Drude model to calculate \( R \) and \( \alpha \). With rapid increase in \( T_e \), \( R \) and \( \alpha \) can dramatically decrease, leading to much more laser energy deposition in the film than that described by the constant \( R \) and \( \alpha \) at RT. For a metal target heated by ultrashort-pulsed lasers with fluence exceeding melting threshold, temperature-dependent \( R \) and \( \alpha \) should be adopted for modeling the thermal response. The present model simulation is only suitable for ultrashort laser pulses with wavelength in 500–1000 nm.