Reflectivity of Intense Femtosecond Laser Pulses from a Simple Metal

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Femtosecond laser heating of a solid offers the possibility of producing plasmas with near-solid densities. Many reflectivity measurements have been made on such plasmas to probe their electrical conductivity. In particular, the experiment of Milchberg, Freeman, and Davey has been interpreted as showing a significant discrepancy between theoretical and measured conductivities. Here we present an alternative interpretation based on careful analysis of hydrodynamic expansion and the nonlocal interaction of the incident laser field. We find the experimental data to be largely consistent with theoretical models. This finding is also supported by the experiment of Fedosejevs *et al.*

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Recent laser experiments have allowed one to explore the transition of electrical conductivity from a metal-like behavior of decreasing with temperature to a plasmalike behavior of increasing with temperature. Since reflectivity is strongly dependent on electrical conductivity through the dielectric function, it is often used as a probe. The first such measurement was reported on dense plasmas produced in the release isentropes of shocked aluminum [1]. The result provided a preliminary validation of a dense plasma conductivity model [2] although the interpretation required detailed modeling of the plasma gradient using a hydrodynamic code.

With the advent of intense femtosecond lasers, the possibility of producing a near-solid-density plasma with a very short gradient scale length has stimulated a new type of experiment [3–8]. Because of limited plasma expansion during the heating pulse, it was thought that the reflectivity of femtosecond lasers can be calculated by considering an electromagnetic wave propagating through a plasma with a steady-state gradient which is assumed to be linear [4,7], exponential [4,5,7] or given by a power law [8]. Its scale length is either estimated from Doppler-shift measurements [6] or left as a free parameter [4,5,7]. The plasma dielectric function is given by the Drude model in which the electron-ion collision frequency is assumed to scale linearly as density. The collision frequency is determined through the dc electrical conductivity. Among these studies, of particular importance is the work of Milchberg, Freeman, and Davey [3], in which they attempted to relate the observed reflectivity directly to the resistivity of a plasma at solid density and at a temperature inferred from the observed Doppler shift. The result showed severe deviations [9] from an existing dense plasma conductivity model [2]. This has stimulated strong experimental interest [4,5] as well as developments of theoretical models [9,10].

However, the apparent controversy hinges heavily on the interpretation of the reflectivity measurement. Given the need for a finite gradient scale length to explain the reflectivity data [3–8] and the extremely transient nature of the heating pulse, the assumption of a constant gradient scale length generated by a steady-state ablative flow is questionable. The much stronger dependence of resonance absorption on gradient scale length than plasma collision frequency for P-polarized light [11] further renders the choice of scale length crucial. In fact, for a steep plasma gradient, reflection of electromagnetic waves occurs over an extended region covering a wide range of densities and temperatures which also vary with time. To elucidate these important issues, we have performed a numerical study of the reflectivity of intense femtosecond laser pulses from aluminum in which the propagation of electromagnetic waves and the dynamics of the plasma are treated self-consistently in a hydrodynamic code. Interestingly, reasonable agreement is found between experimental data [3,4] and results of the simulations using several existing conductivity models [2,10,12].

In numerical calculation, we consider a laser pulse incident from vacuum onto a slab target. The deposition of the laser radiation and the dynamics of the resulting plasma are treated self-consistently by solving the Helmholtz

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equations for electromagnetic waves [13] and the fluid equations for conservation of mass, momentum, and energy in a one-dimensional hydrodynamic code [14]. The initial cold solid and the subsequent hot plasma are characterized by a complex dielectric function $\varepsilon(z,t)=1+i4\pi\times\sigma(\omega)/\omega$ where $\sigma(\omega)$ is the electrical conductivity. At each time step in the calculation, the Helmholtz equations are solved numerically to satisfy boundary conditions corresponding to an incident and a reflected wave in the vacuum, and an evanescent wave inside the target. This yields the reflectivity as well as the complex electric field amplitude from which the energy deposition rate $\langle \mathbf{E}\cdot\mathbf{J}\rangle=\mathrm{Re}(\sigma)|\mathbf{E}|^2/2$ is calculated and incorporated into the hydrodynamic equations as a heat source.

The frequency-dependent electrical conductivity is expressed in the Drude approximation as $\sigma(\omega)=\omega_p^2/4\pi(\nu_{ei}-i\omega)$. Here, $\omega_p^2=4\pi Z^* n_i e^2/m$ is the plasma frequency, Z^* the ionization state, n_i the ion density, m the electron mass, and e the electron charge. The electron-ion collision rate ν_{ei} is given by $Z^* n_i e^2/m\sigma_0$, where the dc conductivity σ_0 is obtained from the calculations of Rinker [12], Lee and More [2], or Perrot and Dharma-wardana [10]. Details of these models have been described elsewhere. The ionization state Z^* is obtained from a screened hydrogenic model [15] in the work of Lee and More and from an atom in jellium model [16] in the work of Rinker. It is calculated self-consistently in Perrot and Dharma-wardana's model.

The above definition of a local dielectric function is generally satisfactory for the treatment of S-polarized light. However, for P-polarized light resonance absorption deposits energy in a region limited by the density gradient scale length at the critical density layer, which can be extremely short in a solid heated by femtosecond laser pulse. Rapid expansion of the heated plasma layer in the resonance region causes the formation of a steepened density step between an underdense and an overdense plasma. This can lead to resonance absorption occurring over an increasingly smaller spatial scale which is ultimately dictated by the mesh size chosen in the simulation. To avoid such an unphysical limit, it becomes necessary to invoke a "nonlocal" prescription for the dielectric function,

$$\varepsilon^*(z)=\frac{1}{\lambda_s}\int_{-\lambda_s/2}^{\lambda_s/2}\varepsilon(z+z')dz'.$$

This yields a dielectric function spatially averaged over a minimum characteristic scale length λ_s. Since screening of electric field can only occur in a spatial scale greater than the degeneracy-corrected Debye length λ_D [4] or the interatomic distance r_0, λ_s is taken to be the greater of these two lengths.

To take into account the effect of non-Maxwellian electron velocity distribution on inverse bremsstrahlung absorption at high irradiances, the Langdon correction on opacity [17] has also been included in the calculation by modifying the electron-ion collision frequency accordingly. For femtosecond laser heating, this effect is less important since absorption occurs mainly at critical and supercritical densities where the collision frequencies are comparable to the laser frequency.

For the hydrodynamic calculation, the equation of state is based on the SESAME tabulated data [18]. The thermal conductivity is calculated using the model of Lee and More [2]. When the diffusive heat flux is less than the free-streaming heat flux, the calculation assumes no flux saturation. When the diffusive heat flux becomes comparable to the free-streaming heat flux, a harmonic-mean flux limiter model [19] is used.

For plasmas heated by femtosecond laser pulses, the electron temperature would be much higher than the ion temperature. However, in the absence of any theoretical or experimental data on equation of state and electron-ion coupling in a nonequilibrium dense plasma, we have assumed that the electrons and the ions are in thermodynamic equilibrium. Since the hydrodynamic motion of the plasma is governed by the kinetic energy of the electrons and the inertia of the ions, it is not expected to be sensitive to the ion temperature. The effect of nonequilibrium between electrons and ions on electrical conductivity has been explored [20] but complete calculations of transport properties remained outside the scope of the present work.

Since the $I\lambda^2$ value (I and λ are, respectively, the intensity and wavelength of the laser radiation) of concern is limited to 1.6×10^{14} W$\,\mu$m^2/cm^2, other high intensity effects such as profile modification by ponderomotive force [21], parametric instabilities [22], hot electron production and transport, spatial dispersion, and wave breaking [23] are ignored. Radiation transport and vacuum heating [24] are also not treated in the calculations.

As an example of the numerical study, Fig. 1 shows results of the simulation corresponding to the experiment of Milchberg, Freeman, and Davey [3] at an irradiance of 10^{14} W/cm^2. Lee and More's conductivity model is used. The nonsteady nature of the plasma expansion is illustrated by the temporal variation in the density gradient scale length at the critical density layer. The assumption of a single scale length throughout the duration of the laser pulse is clearly invalid. The change in the plasma also leads to a time-varying reflectivity. For S polarization, the reflectivity drops at the onset of the laser pulse, characteristic of a solidlike behavior of decreasing conductivity with increasing temperature of the target. With further heating, this changes into a plasmalike behavior of increasing conductivity and therefore reflectivity with temperature. For P polarization, the initial decrease in reflectivity is followed by a further reduction when resonance absorption becomes significant. Such details are not revealed in the experiment which yields only the time-integrated reflectivity.

Since reflectivity results from the interaction of the electromagnetic wave with the plasma gradient as a whole, it cannot in general be related to a single plasma

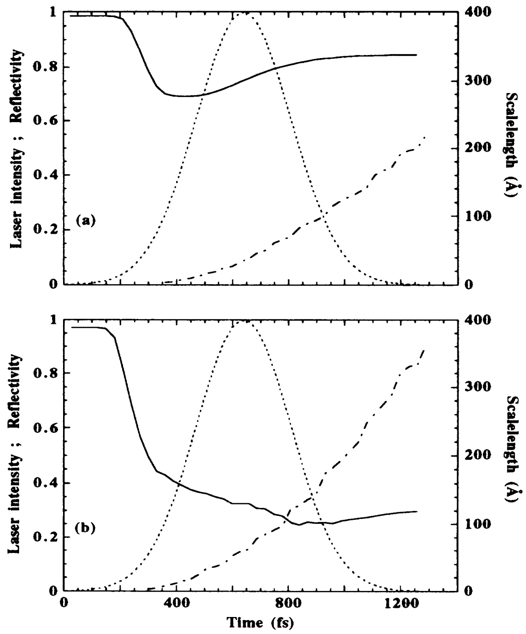

FIG. 1. Temporal variations in plasma reflectivity (solid line) and density gradient scale length at critical density (dot-dashed line) for (a) S- and (b) P-polarized, 400 fs, 308 nm radiation at 10^{14} W/cm^2 and 45° incidence. The incident laser pulse (dotted line) is also shown.

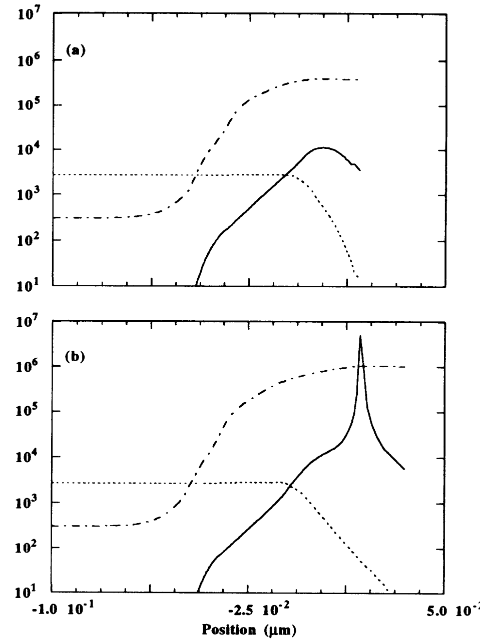

FIG. 2. Profiles of plasma density in 10^{-3} g/cm^3 (dotted line), temperature in K (dot-dashed line), and absorbed power in 10^{15} W/g (solid line) at the peak of the laser pulse for the same condition as Fig. 1. The initial position of the target surface is at 0 μm.

layer. The extent of the plasma region and hence the range of plasma densities and temperatures responsible for the observed reflectivity can be seen from the laser absorption profile in the target (Fig. 2). For S polarization, absorption occurs mainly in the overdense region. In the case of P polarization, a dominant spike characteristic of resonance absorption is found near the critical density layer. Evidently, the electromagnetic wave-plasma interaction occurs over a wide range of plasma conditions. The interaction becomes even more delocalized at lower irradiances when the plasma gradient remains steeper. Thus, the absorption and the reflection process cannot be readily characterized by single-valued plasma parameters.

Figure 3 shows the irradiance dependence of the reflectivity corresponding to the experiment of Milchberg, Freeman, and Davey [3]. For P polarization, the overall agreement between calculation and measurement is quite good for all three electrical conductivity models particularly for the model of Perrot and Dharma-wardana. This contradicts the severe discrepancy [9] suggested by the plasma resistivity inferred by Milchberg, Freeman, and Davey. For S polarization, Perrot and Dharma-wardana's model again yields the closest agreement with data although some discrepancies remained for irradiances above 10^{14} W/cm^2. This can be attributed to residual deficiencies in the conductivity model. It may also be due to stronger nonthermal equilibrium effects (unequal electron and ion temperatures) at the higher irradiances.

As a further test of our simulation, calculations have been made to probe the angular dependence of the reflectivity corresponding to the experiment of Fedosejevs *et al.* at an irradiance of 10^{14} W/cm^2 [2]. The results are presented in Fig. 4. In the calculation, the change in the irradiance with angle of incidence θ has been taken into account by assuming a $\cos\theta$ dependence. The effect of polarization mix (93% of the total laser intensity resides in the desired polarization direction) is also included. In this case, the electrical conductivity model of Lee and More yields better agreements between theory and experiment. All models show that maximum resonance ab-

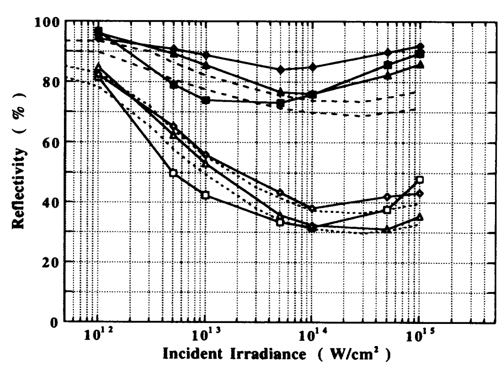

FIG. 3. Comparison of simulation to the experiment of Milchberg, Freeman, and Davey [3]. S/P polarization: upper and lower bounds of experimental data (dashed/dotted lines), calculations using Lee and More's conductivity (solid/open squares), Perrot and Dharma-wardana's model (solid/open triangles), and Rinker's model (solid/open diamonds).

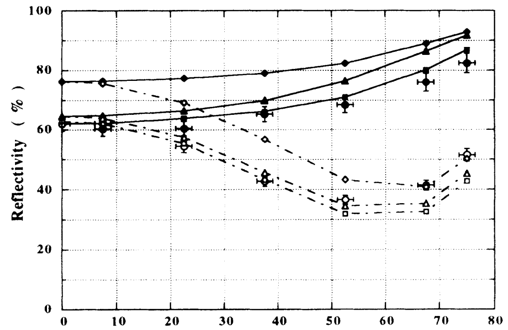

FIG. 4. Comparison of simulation to the experiment of Fedosejevs *et al.* [4]. S/P polarization: experimental data (solid/open circles), calculations using Lee and More's conductivity (solid/open squares), Perrot and Dharma-wardana's model (solid/open triangles), and Rinker's model (solid/open diamonds).

sorption occurs at about 60°, in good agreement with observation. This would support the accuracy in our treatment of the plasma expansion.

For S polarization, the diffusive heat flux is small compared with the free-streaming heat flux for all conditions studied here. This is not unexpected since absorption occurs mainly in the solid material and the scale length of the heat front is governed by the penetration depth of the evanescent electromagnetic wave (Fig. 2), which is usually much longer than the electron mean free path at solid density. For P polarization, resonance absorption produces a steep temperature gradient at the critical-density region (Fig. 2). The diffusive heat flux becomes comparable to the free-streaming flux for irradiances exceeding 10^{14} W/cm^2. For such high irradiances, the calculations were performed using a flux limiter of 0.6 (Fig. 3). It may be noted that the reflectivity increases with lower flux limits. Reduction of thermal transport leads to higher temperatures and expansion velocities in the coronal plasma, which in turn leads to steepening of the plasma gradient and modification of the resonance absorption process. However, the experimental data appear to suggest a flux limit close to 0.6.

In conclusion, we have presented results of the first hydrodynamic calculation of the reflectivity of intense femtosecond laser pulses from a simple metal, which treats self-consistently the propagation of an electromagnetic wave in a plasma gradient as well as the expansion of the plasma. A novel feature of the model is the use of a nonlocal dielectric function prescription with an arbitrary but reasonable minimum cutoff in its scale length. Simulations using an *ab initio* conductivity model [10] based on density functional theory show the best agreement with experimental observations. It appears that proper treatment of the plasma dynamics has explained the controversy regarding the failure of such conductivity models in the region of maximum resistivity [9,10]. In fact, this has clearly illustrated the need of modeling plasma expansion even for laser pulses of few hundred femtoseconds. Because of the very short duration of the laser pulses, it would be interesting to include the treatment of nonequilibrium between electrons and ions in future calculations.

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