Efficient coupling of high-intensity subpicosecond laser pulses into solids

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We demonstrate a new technique for enhancing the absorption of high-intensity, ultrashort-duration laser pulses by solids. Targets consisting of gold gratings and gold clusters were found to absorb greater than 90% of the incident high-intensity laser light. This is in contrast to less than 10% absorption by flat surfaces. As a result of this strong coupling of the laser to a high-density plasma, conversion efficiency of laser energy to x rays of greater than 1% was observed for x rays above 1 keV. Efficiency of nearly 25% was observed for emissions greater than 30 eV. These conversion efficiencies are more than an order of magnitude greater than those measured from flat targets.

High-density plasmas generated by intense, femtosecond lasers have been demonstrated as sources of subpicosecond x-ray pulses in the soft x-ray region.[1,2] However, a major limitation to their general use has been the relatively low laser energy to x-ray energy conversion efficiency observed to date ($<1\%$). This low efficiency is primarily due to the inefficient coupling of the laser energy into the solid, and also the relatively low plasma temperatures produced.

The inefficient laser-to-solid coupling in the case of a flat target can be understood using a simple model.[3,4] When a flat solid surface is illuminated with a high-intensity ultrashort laser pulse, the solid rapidly evolves into a high conductivity plasma. For a flat silicon wafer irradiated with a 150 fs laser pulse at an incident intensity of 10^{16} W cm^{-2}, a plasma with an estimated peak electron temperature of 400 eV is created at the surface. The complex refractive index is calculated approximately at $N=n+ik\approx4.5+i10.5$. The normal incidence reflectivity R is then given by $|(N-1)/(N+1)|^2$. Thus, for the high values of the n and k characteristics of a dense plasma, the peak reflectivity is calculated to be 90%, and the laser light is reflected from a mirrorlike, hot-metal surface.[2] This prediction is not changed by the presence of a thin layer of low density material ($\ll\lambda$) formed in front of the solid due to plasma expansion during the excitation pulse.[5] We note however, that the presence of a significant density gradient in front of the target can increase short pulse absorption.[1,6–8]

In an attempt to increase absorption of the laser by *solid* density material, we studied two types of structured targets which exhibit low reflectivity.[9,10] The first was a grating structure and the second consisted of a low average density matrix of solid metal clusters. The grating targets used have periodicity less than the laser wavelength of 6200 Å to avoid energy loss in a diffracted beam, and have groove widths sufficiently wide ($\geqslant$1000 Å) so that they do not fill with low-density blowoff vapor during the laser pulse. The gratings were fabricated using both electron-beam lithography and holographic exposure techniques and were used at normal incidence. Gratings with a periodicity of 2400 and 3000 Å were both studied, with the fill factor (fraction of the period occupied by the ridge) ranging from 0.33 to 0.66. The reflected light from the gratings was specular and could be accurately measured.

Figure 1(a) shows the incident laser reflectivity as a function of grating depth, at an incident power density of 10^{16} W cm^{-2} and for incident laser polarization perpendicular to the grooves. As is seen in Fig. 1, for groove depths greater than $\approx$1000 Å the laser light is mostly absorbed. In contrast, for laser light polarized parallel to the grooves, the reflectivity was always $\geqslant$50%. The gratings studied were made from silicon shown in Fig. 2 or photoresist, and were sometimes overcoated with gold. The measured reflectivity was relatively independent of target composition, fill factors and groove width. We calculate that the absorption per unit surface area increases by up to a factor of 3 in these targets.

Since the grating periodicity was of order $\lambda/2$, a long wavelength, mean field approximation cannot be used to calculate the effective refractive index of the structure.[11] However, the grating structure can be modeled as a set of parallel lossy waveguides. For radiation polarized parallel to the grooves, the guide is cutoff and the structure is reflective. For radiation polarized perpendicular to the grooves, only a fraction of the incident radiation is reflected from the top surface[12] and most of the radiation is coupled into a propagating TE mode in the grooves. Attenuation in this structure is given by[13]

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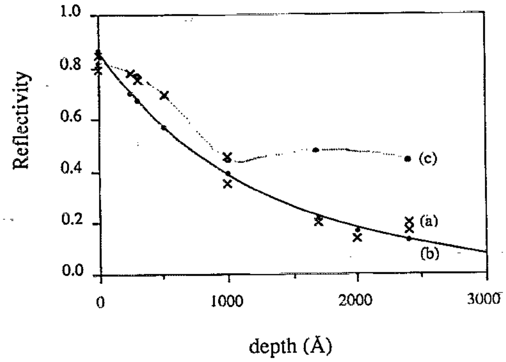

FIG. 1. Reflectivity of grating targets as a function of groove depth at a laser intensity of 10^{16} W cm^{-2}: (a) experimental values (crosses), (b) approximate waveguide calculation (dark circles), (c) coupled wave calculation (grey circles). The curves are as a guide to the eye only.

$$\alpha=\frac{1}{a}\sqrt{\frac{2\omega m\epsilon_0}{ne^2\tau}},$$

where α is the power loss coefficient, a is the groove width, ω is the laser frequency, ϵ_0 is the dielectric constant, n, e, and m are the electron density, charge, and mass, respectively, and τ is the electron-ion collision time. When $\omega=3\times10^{15}$ s^{-1} and $a=1200$ Å, and assuming approximate values for a hot, dense plasma of $n=1\times10^{23}$ cm^{-3} and $\tau\approx1\times10^{-16}$ s, we obtain $\alpha^{-1}\leqslant3000$ Å. This implies a groove depth of 1500 Å for $1/e$ attenuation of the laser power, assuming that light is reflected from the back surface of the structure, which is in reasonable agreement with the experimental data as seen in Fig.1(b).

The simple model discussed above does not accurately model the electric field distribution and interference effects within the periodic structure. Therefore, a more rigorous coupled-wave analysis[11] for the metal coated grating was performed. The predictions are sensitive to the assumed structure and temperature profile; here the grooves are assumed to be perfectly rectangular, and the temperature is assumed uniform throughout the wall material. The reflectivity predicted by this model for the high-temperature case is shown in Fig. 1(c).

A second type of target which exhibits low reflectivity is made up of gold clusters, and is often called gold smoke or gold black. This matrix consists of chains of clusters of ≈50 Å particle size, which form a low-average-density structure (typically $\approx0.3\%$ of solid density), but a high local density. Gold black is fabricated[14] by evaporating gold in background of several Torr of gas which causes the gold to cluster and form a microscopic fractal structure.[15,16] It is strongly absorbing throughout the visible.[17–19] Figure 3 shows an electron microscope picture of the gold cluster material.

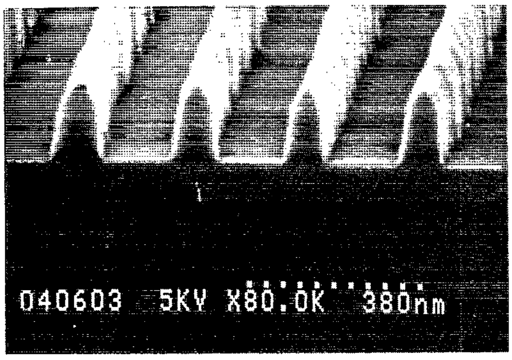

FIG. 2. Electron microscope picture of a silicon grating target, with period 2400 Å and groove depth ≈1700 Å.

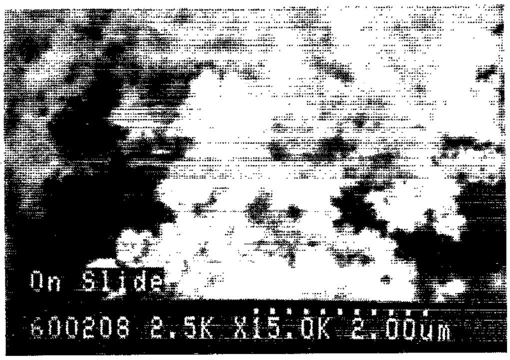

FIG. 3. Electron microscope picture of a gold cluster target.

By considering Mie scattering[20] from a spherical, solid gold particle with diameter d, we can derive a simplified model for absorption by this structure. In the long wavelength approximation ($d/\lambda\ll1$) the field inside the particle, E_{in}, can be expressed in terms of the applied field E_0, as $E_{in}/E_0=3/(\epsilon_m+2)$, where $\epsilon_m=\epsilon_b-\omega_p^2/(\omega^2+i\omega/\tau)$. Here ϵ_m is the dielectric function of the material of the particle, ϵ_b is the contribution due to interband transitions, ω_p is the plasma frequency of gold, ω is the laser frequency, and τ is the collision time. At the frequency for which $\mathrm{Re}(\epsilon_m)=-2$, there is a resonant surface plasma oscillation in the sphere. This corresponds to a shift of the plasma resonance towards the visible region of the spectrum, from the ultraviolet plasma resonance in bulk gold at $\omega_p=8.6$ eV. For gold at room temperature, $\epsilon_b\approx7.5$,[21] and thus the above equation predicts the surface plasma resonance of gold particles to be at approximately 2.8 eV; the measured optical constants of gold[21] would predict it to occur at 2.5 eV. Experimentally, our gold cluster targets typically have absorption resonances between 2 and 1.6 eV, which is similar to what has been observed by other groups.[16,22–24] Therefore initially our 2 eV photons correspond to near-resonant excitation of the gold clusters. Once hot, the interband contribution to the dielectric function vanishes, and the bulk plasma frequency will change as a result of expansion and ionization of the cluster. These effects will tend to move the surface plasma resonance towards the ultraviolet.

An exact calculation of the position and width of the plasma resonance must take into account the cluster shape and size,[25] the electrical contacts between individual clusters, the reduction in electron collision time (from 30 fs in bulk gold, to ≈3 fs in the 50 Å gold clusters), and the local fractal structure[15] (a correlated collection of interacting multipoles). As a result, as was the case of the grating targets, for accurate calculations a mean-field, average-index model is not expected to be accurate. We can however, neglect interactions and estimate the absorption coefficient for a collection of small spheres with average density f compared with a solid by[22]

$$\alpha = \frac{18\pi f}{\lambda} \frac{\epsilon_2}{(\epsilon_1+2)^2+\epsilon_2^2}.$$

where ϵ_1 is the real part and ϵ_2 is the imaginary part of ϵ_m and $f \approx 3\times 10^{-3}$. On resonance ($\epsilon_1=-2$, $\epsilon_2=3$), this expression predicts an absorption depth $\alpha^{-1} \approx 10\ \mu\text{m}$.

During the laser heating process, the structure will begin to homogenize due to expansion. We estimate the time required to homogenize the material by considering the expansion of a 50 Å gold cluster, of initial temperature 300 eV, to a 400 Å diam sphere. This time is ≈400 fs, so that it is unlikely that our structures fill in completely during the excitation pulse. Substantial cooling due to expansion in three dimensions is possible, which may lower the temperature by a factor of ≈20 during the initial picosecond after excitation.[26] Thereafter, cooling will be slower due to reduced gradients.

From the predicted scaling of plasma temperature and the resulting x-ray yield with absorbed laser energy,[2,27] we estimate both an increase in both the temperature (≈2) and the total x-ray yield (≈10) for the grating and cluster targets compared to the flat targets. Absolute x-ray yields from both target types were experimentally measured using a filtered x-ray diode (United Detector XUV005HS), placed 35 cm from the plasma, using the manufacturer-supplied calibration factor. For gold black, we measured a laser to x-ray conversion efficiency approaching 25% for x rays above 30 eV, with ≈1% of the x rays at energies above 1 keV. For gold coated gratings, we observed a conversion efficiency of ≈12%, with ≈0.6% above 1 keV. For flat gold, the measured conversion efficiency was ≈0.9% for x rays above 30 eV.

The x-ray pulse duration from both the grating and cluster targets was measured using a fast x-ray streak camera.[28] We measured instrument-limited x-ray pulsewidths of 2 ps for the high-energy photons (>1 keV) from both targets. The true pulsewidth is expected to be subpicosecond when the instrument response is deconvolved.[28] For low energy x rays (≈100 eV) the measured pulsewidth was somewhat longer (≈5 ps). Gold cluster emissions in this soft x-ray region were approximately a factor of two longer than those of grating targets, possibly due to slower cooling at lower material density after a few picoseconds of expansion. This increase in x-ray pulsewidth may be due to space-charge effects, which are severe for short pulses.

In summary, we have demonstrated a new method of coupling the energy of high-power, short-pulse lasers into solid targets. We measured conversion efficiencies of laser light to short pulse x rays above 1 keV of approximately 1%; this corresponds to a peak x-ray power of 20 MW. The advantages of structured targets include the possibility of increased energy deposition per volume, as well as increased x-ray emissions from these structures.

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