High-Density Plasmas Produced by Ultrafast Laser Pulses

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We describe temporal and spectroscopic measurements of high-density plasmas produced by focusing intense, 160-fsec laser pulses on solids. Soft-x-ray emission with a duration of 2 ± 2 psec is observed up to photon energies of a kilovolt. We observe reduced emission from long-lived spectral lines, indicating the presence of a short-lived, high-density plasma. Reflectivity measurements indicate that absorption of the laser pulse occurs at the surface of the solid before it expands.

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The interaction of intense, subpicosecond laser pulses with solid targets is of current interest both for the study of high-density plasmas and for the production of short-pulse x-ray sources.[1-10] When an intense laser is focused onto the surface of a solid, a high-temperature, x-ray–emitting plasma is produced. If the laser pulse is short, the energy can be absorbed by the electrons in an optical skin depth, leading to rapid ionization before significant ablation of the solid occurs. Rapid cooling of hot electrons and subsequent quenching of the x-ray emission are expected due to rapid thermal conduction into the underlying cold material, electron energy loss to ions, and expansion into the surrounding vacuum.[1,2,8-10]

In this Letter we describe studies of this type of plasma, obtained using a dye laser[11] at 616 nm with a pulse width of 160 fsec, an energy of 5 mJ, and a repetition rate of 10 Hz. The focusing optics was an off-axis parabolic mirror which focused the laser pulses to intensities exceeding 10^{16} W cm^{-2} onto a solid target mounted on a scanning stage. Plasma temperatures of 230 eV were estimated from both the intensities of the emission lines from various ionization stages and reflectivity measurements; this temperature is consistent with predictions of one-dimensional heat-flow models describing the laser-solid interaction.[1,2] Emission from the plasma extends over a broad spectrum up to photon energies of at least 1 keV.

A Kentech x-ray streak camera with a 100-μm slit and an extraction field of 35 kV/cm was used to measure the emission pulse width. The composite photocathode consisted of Lexan (100 nm), aluminum (25 nm), and potassium bromide (150 nm). This photocathode has good quantum efficiency[12] at photon energies exceeding 30 eV. The time resolution and sweep-speed calibration of the streak camera were obtained with two 500-fsec laser pulses at 308 nm separated by a known time delay. These uv pulses had to be focused on the photocathode to obtain a response because of the extremely low photoelectron yield at this wavelength. A charge-coupled-device camera and computer system were used to record the streak-camera output with a precision of 0.3 psec and with a sensitivity of a single photoelectron.

Figure 1 shows typical streak-camera traces of the plasma x-ray emission from a silicon target [Fig. 1(a)], and the response to the uv-laser pulse [Fig. 1(b)]. The full-width at half-maximum intensity pulse widths are 6.0 and 5.6 psec, respectively, and are repeatable to $\pm 10\%$ of these values. Although deconvolution yields an x-ray pulse width of 2 ± 2 psec (assuming Gaussian pulse shapes), it is not strictly correct to deconvolve the measured widths to obtain the true x-ray pulse width. Since the electron energy distribution is not the same for the x-ray and uv excitation, temporal dispersion[10] through the streak camera leads to a different impulse response at each wavelength. For uv excitation, the energy distribution is narrower since the photoelectrons arise from direct ionization of a narrow-band F center.[13] Therefore, deconvolution gives only an *upper limit* for

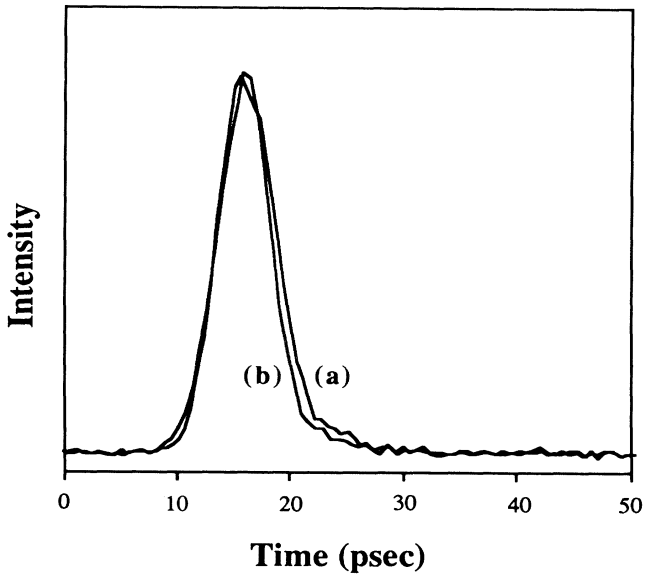

FIG. 1. (a) Streak-camera measurement of the soft-x-ray pulse width from a silicon plasma showing a FWHM of 6.0 psec. (b) Streak-camera response with a 500-fsec, 308-nm pulse incident on the same photocathode as in (a), showing a FWHM of 5.6 psec.

the x-ray pulse width. For silicon, the pulse width was measured both with and without a 100-nm-thick filter of aluminum between the plasma and the photocathode; no change in pulse width was observed with the filter, which eliminated all radiation below 50 eV. In order to investigate radiation in the keV region, a tantalum target was used with an 8-μm-thick beryllium filter to eliminate all radiation below 800 eV. A slightly shorter emission time was observed in this case, probably due to more rapid quenching of the high-energy states giving rise to this radiation. The tantalum target also yielded signals 10 times more intense than silicon for all energy ranges. At very high intensities, space-charge effects in the camera lead to broadening of the measured pulse duration for both x-ray and uv illumination.[14,15]

Very different results were obtained when the short laser pulse was preceded by laser energy on a time scale of a few nanoseconds which resulted from amplified spontaneous emission (ASE) originating in the dye-laser amplifiers.[11] Figure 2(a) shows a 77-psec x-ray decay time from a silicon target obtained when the ASE energy was about 10% of the energy in the short laser pulse. As shown in Fig. 2(b), this reduced to 40 psec when a filter was used to eliminate all radiation below 50 eV. Figure 2(c) shows the short-pulse x-ray emission obtained when the ASE was less than 10^{-4} of the short-pulse energy, for comparison. In all cases the fast rise times are streak-camera limited at 2.7 psec. When the ASE energy was reduced to 5%, the decay time was reduced to 10–20 psec; the threshold for observable x-ray pulse broadening was about 4 μJ of ASE. ASE energy alone did not generate any detectable short-wavelength radiation. It was introduced into the system by the reduction of the number of saturable absorbers in the dye amplifier chain[11] and preceded the short pulse by several nsec.

We interpret the long measured decay time in the presence of ASE as emission coming from a lower-density plasma compared to the case of no ASE. Energy which precedes the short-pulse laser can vaporize the solid and create a region of lower electron density away from the solid surface which will effectively absorb the short-pulse laser at the critical density, where the plasma frequency equals the optical frequency.[3,16] In this lower-density plasma, two- and three-body recombination rates will be reduced; also, reduced thermal conduction and slower expansive cooling will result from the decreased temperature and density gradients due to longer scale lengths. These conditions lead to an expected increase in the x-ray-emission decay time, which is consistent with our observations.

Additional evidence for high-density plasma formation was obtained from time-integrated spectroscopy experiments. Emission on Si V ($2p^6$-$2p^5 3s$) transitions at 11.79 and 11.90 nm, from singlet and triplet excited states, respectively, were compared for various plasma

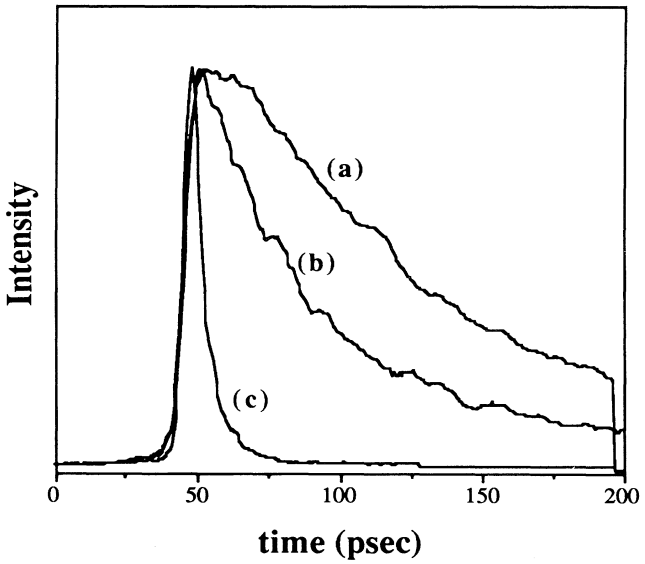

FIG. 2. Soft-x-ray pulse widths obtained from a silicon plasma; peak intensities have been normalized. (a) Short-pulse excitation with 10% ASE energy present. (b) As in (a), but filtered to show response above 50 eV only (intensity multiplied by 4). (c) Short-pulse excitation with less that 10^{-4} ASE energy background (intensity multiplied by 3).

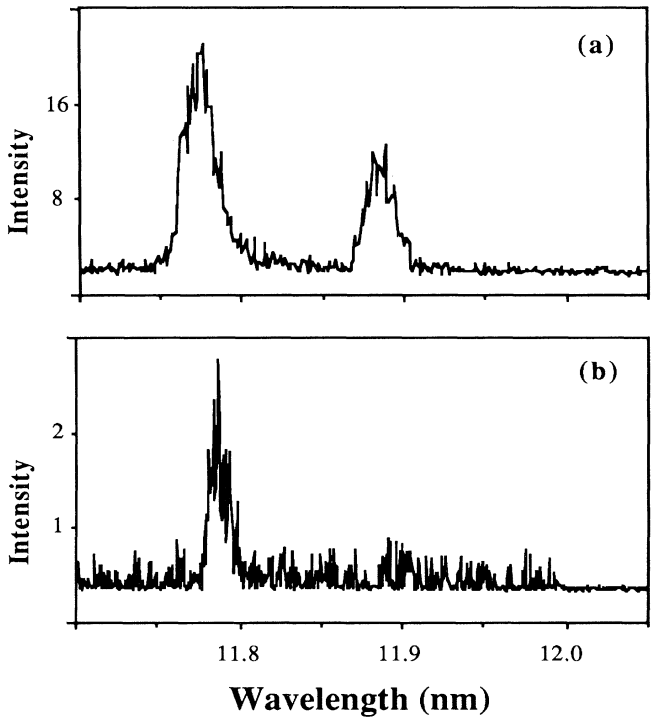

FIG. 3. Plasma emission at the Si V ($2p^6\,{}^1S$-$2p^5 3s\,{}^1P,{}^3P$) transitions at 11.79 and 11.9 nm. (a) Short-pulse laser illumination with 10% ASE energy background. (b) Short-pulse laser illumination only.

excitation conditions. ASE energy alone focused on the target yields an emission intensity ratio of 1:1 for the singlet and triplet transitions, which is identical to observations from low-density plasma discharges.[17] When the short laser pulse is simultaneously incident on the target with the ASE background, as shown in Fig. 3(a), the emission ratio is 2:1. This ratio increases further in the absence of the ASE background, as shown in Fig. 3(b). Similar behavior is observed from the Al IV ($2p^6$-$2p^5 3s$) transitions at 16.01, and 16.17 nm. In silicon (aluminum), the radiative lifetime of the singlet excited state is 33 psec (59 psec) while the triplet-state lifetime is 300 psec (770 psec).[17] The increase in the singlet-triplet ratio under short-pulse illumination is consistent with collisional mixing of excited states at high densities and rapid quenching of the plasma. At high densities, the emission ratio is expected to approach 9:1 as the populations of the states approach their high-temperature equilibrium values and radiate in proportion to their respective emission rates. Figure 3(b) indicates a ratio greater than 5:1 and is therefore consistent with emission from a high-density short-lived plasma.

Figure 4 shows the reflectivity of the short-pulse laser from the silicon target as a function of laser fluence (measured at an incident angle of 5° to the normal). The results can be compared with model calculations[1] of reflectivity of the solid versus electron temperature, shown in Fig. 5. At low temperatures, reflectivity is computed from data[18] on room-temperature and liquid silicon. In the crosshatched region the reflectivity was not calculated because the plasma model is not expected to apply. At high temperatures, the calculations assume a Drude model for the dielectric constant[19] of the ionized material with equilibrium ionization conditions computed as a function of electron temperature. Figure 4(a) shows the measured reflectivity (R) of silicon as it evolves from a room-temperature solid at low laser fluence, with $R=35\%$, to a high-temperature, partially ionized plasma, with $R=75\%$. At a laser fluence of 300 J cm^{-2}, the reflectivity data are consistent with the model at temperatures of approximately 230 eV when we spatially and temporally average over the laser pulse. The reflected short-pulse laser beam is specular and image preserving even at the highest intensities. Previous measurements[20,21] have determined the reflectivity of silicon up to fluences of about 2 J cm^{-2} and are in agreement with Fig. 4(a). Figure 4(b) shows the reflectivity of a typical long-pulse (7 ns), 532-nm laser pulse. In this case, the reflectivity is significantly lower for a given energy fluence due to efficient coupling between the laser and the lower-density material formed by vaporization of the solid during the rising edge of the pulse.[3,16] In addition, the reflected portion of the 7-ns pulse is completely diffuse with no retention of beam structure. The reflectivity in the long-pulse experiment was measured with a large-area lens to sample a known solid angle, from which the total reflectivity could be deduced since the reflected beam was reasonably uniform over the half-sphere. Both input and output beams were sampled on each shot and the measurements were repeatable to within 10%.

In summary, we have measured soft-x-ray emission from a plasma produced by a high-intensity 160-fsec laser to be less than 2 psec in duration. We have demonstrated that for plasmas heated by a short-pulse laser in conjunction with a long-pulse background the x-ray duration can be more than an order of magnitude longer in time. Additionally, spectroscopic experiments indicate

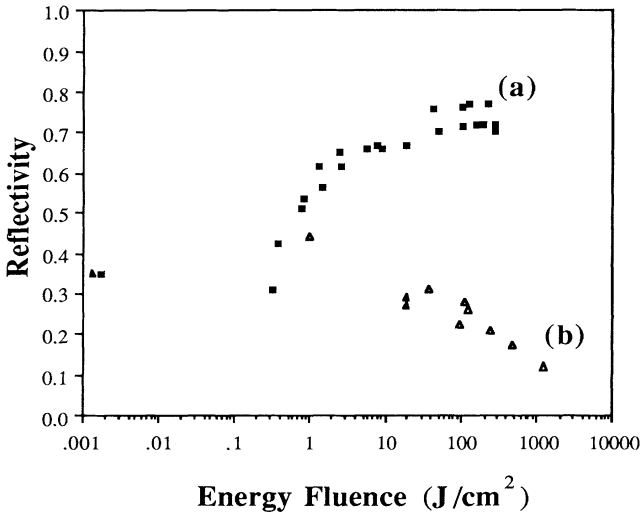

FIG. 4. Experimental values for the reflectivity of silicon as a function of energy fluence on target. (a) Short-pulse laser illumination. (b) 7-nsec-long laser pulse illumination.

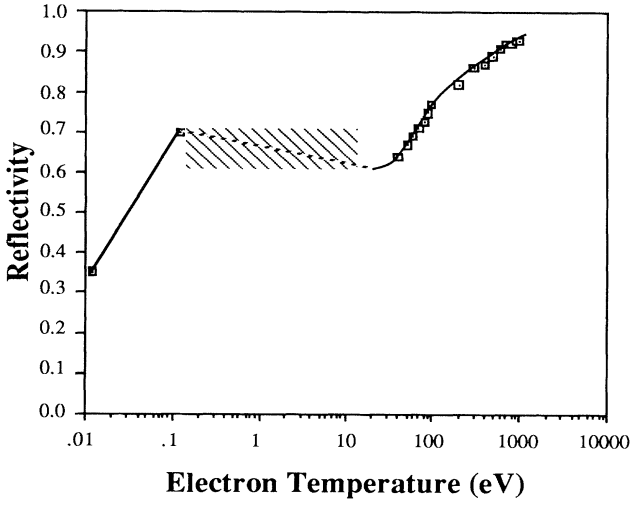

FIG. 5. Model values for the reflectivity of a silicon target as a function of temperature with use of a Drude model of a solid-density, ionized plasma.

that a rapidly quenched, high-density plasma is formed under short-pulse illumination conditions. Finally, reflectivity measurements are consistent with absorption of the short-pulse laser at solid densities, in contrast to longer-pulse illumination for which the coupling is into a preformed, lower-density plasma.

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