Control of Bright Picosecond X-Ray Emission from Intense Subpicosecond Laser-Plasma Interactions

J. Workman, A. Maksimchuk, X. Liu, U. Ellenberger, J. S. Coe, C.-Y. Chien, and D. Umstadter
Center for Ultrafast Optical Science, University of Michigan, Ann Arbor, Michigan 48109-2099
(Received 23 February 1995)

Using temporally and spectrally resolved diagnostics, we show that the pulse duration of laser-produced soft x rays emitted from solid targets can be controlled, permitting a reduction to as short as a few picoseconds. To enable this control, only a single parameter must be adjusted, namely, the intensity of the high-contrast ultrashort laser pulse (400 fs). These results are found to be in qualitative agreement with a simple model of radiation from a collisionally dominated atomic system.

PACS numbers: 42.60.Gd, 07.85.Fv, 32.30.Rj, 52.25.Nr

The interaction of a high-intensity high-contrast ultrashort laser pulse with a solid target generates unique conditions for ultrashort duration x-ray generation [1]. Because relatively little expansion occurs during the laser pulse, the density scale length remains much less than the laser wavelength [2]. This makes possible the direct deposition of a significant amount of the laser energy at near-solid density. Rapid changes in the plasma parameters—caused by their steep gradients—determine the subsequent evolution of the x-ray emission.

Understanding and ultimately controlling laser produced ultrashort x-ray sources is essential for their application, which includes x-ray laser schemes, probing hot-dense matter and time-resolved diffraction, spectroscopy, or microscopy studies of transient physical, chemical, or biological phenomena [3]. Mechanisms for controlling x-ray pulse duration have been suggested theoretically, including the use of mixed species targets to increase cooling rates [4] and the use of electron temperature to control recombination emission [5]. However, up until this point, only one parameter has been demonstrated experimentally to control the ultrashort x-ray pulse duration, namely, the density scale length [6–8]. It was demonstrated that the x-ray pulse duration could be decreased [6] by increasing the laser contrast, which effectively decreases the density scale length. In other studies [7,8], the density scale length was increased using a prepulse, with the desired effect of increasing the absorption of the main laser pulse—and thus x-ray emission—but with the disadvantage of increasing the x-ray pulse duration.

In this Letter we demonstrate, for the first time, the control of x-ray pulse duration by adjusting the incident laser intensity, and thus the electron temperature, leaving the density scale length relatively unchanged. Specifically, we show that a lower peak electron temperature leads to a shorter pulse of x rays, which are shown to be as short as a few picoseconds in duration. We study soft x-ray emission, which can be measured over a large range of electron temperatures and thus laser intensities, instead of high-energy keV emission, which—because of it's lower flux—can only be measured at the highest laser intensities [9]. Our experimental results are qualitatively explained with a simple analytic model of radiation from a collisionally dominated atomic system. These measurements should provide a benchmark for more complex numerical models of these unique laser-plasma interactions.

In performing the experiment, we used a 400-fs terawatt Nd:glass laser system based on chirped pulse amplification. The intensity contrast ratio of the peak of the fundamental 1.06-μm laser light to the amplified spontaneous emission (ASE) pedestal, measured to be 5×10^5, was increased to 10^{10} by frequency doubling. An off-axis parabolic mirror was used to focus the S-polarized 2ω laser radiation onto a solid target at normal incidence. A minimum spot size of 15 μm with an energy of about 300 mJ was achieved, corresponding to a maximum intensity in the second harmonic light of 5×10^{17} W/cm^2. Targets included 4-μm-thick aluminum and gold deposited onto silicon wafers. In order to decrease the incident intensity on the target, the laser spot size was defocused while keeping the total laser energy constant.

The soft x-ray emission was spectrally dispersed using an imaging flat-field grazing-incidence variable-spaced grating spectrometer located at an angle of 45° to the target normal. With a spectrometer entrance slit width of 200 μm, we obtained a spectral resolution $\lambda/\Delta\lambda$ of about 300 at 50 Å. Time-resolved spectra were obtained in a single shot, using an x-ray streak camera with a potassium bromide photocathode supported by a Lexan film, coupled to the spectrometer. The temporal resolution was 5 ps. A 20-μm x-ray pinhole camera, filtered with 25 μm of Be and 6000 Å of Al, was used to monitor the laser spot size and emission region. Two p-i-n diodes filtered with 50 and 100 μm of Be were used to monitor relative keV x-ray emission levels and to monitor the reproducibility of each interaction. A calibrated calorimeter was used to monitor the laser energy of each shot. To obtain quantitative conversion efficiencies of the laser radiation into x rays, in the range 1.5–5 keV, DEF film was used with steps of Be filters of different thicknesses and with known characteristic absorption curves [10].

 0031-9007/95/75(12)/2324(4)$06.00

The dependence of x-ray pulse duration on laser intensity is demonstrated in Fig. 1, which shows single-shot aluminum x-ray emission for three different laser intensities, ranging from 2.0×10^{17} down to 2.0×10^{15} W/cm^2. X-ray emission in this region of 45 to 70 Å is dominated predominantly by Al XI and Al X ions, where the strongest feature observed is the Al XI $2p$-$3d$ emission line at 52 Å. The decrease in x-ray pulse duration over the entire spectrum with decreasing laser intensity is clearly demonstrated in this figure. At the lowest laser intensity one can see the emission from lower ionization stages in the 55 to 60 Å wavelength region.

The change in the ionization balance is seen in Fig. 2, which shows the spectral traces of Fig. 1 at the peak of the emission for each laser intensity [11]. Indicated in the figure are the Al XI $2p$-$3d$ and $2s$-$3p$ transitions as well as the Al X $2s2p$-$2s3d$ and $2p^2$-$2p3d$ transitions. One can see clearly that the Al X ionization stage becomes stronger with respect to the Al XI ionization stage as the laser intensity is decreased, indicating a corresponding decrease in peak electron temperature. The Stark broadening of the line emission, seen in Fig. 2, demonstrates that the emission originates from high density in each case. (We measured a spectral resolution of approximately 0.2 Å in a separate experiment using long pulse (nanosecond) illumination of both carbon and beryllium. As can be seen in Fig. 2, the broadening is on the order of 1 Å or greater.)

Figure 3 shows a comparison of amplitude-normalized temporal traces from Fig. 1 of the Al XI $2p$-$3d$ emission line ($\lambda = 52.4$ Å) obtained for 2ω irradiation for a range of laser intensities. Again, one clearly sees the pronounced decrease of the x-ray pulse duration as the laser intensity is decreased. The rise time is short in all cases as expected from the rapid heating. (The starting positions of the plots relative to the laser pulse and each other are unknown.) The fall time is observed to be shorter and the slope of the decay much steeper in the lowest-laser-intensity case. Similar results were obtained for the Al XI $2s$-$3p$ transition.

Figure 4 shows the comparison of measured x-ray pulse duration as a function of laser intensity for two different materials, aluminum and gold. The averaged experimental aluminum pulse widths at $\lambda = 52.4$ Å are shown as crosses, while the gold data, also at $\lambda = 52.4$ Å is represented by the open diamonds. The data were averaged over three to five laser shots with the standard deviation of the pulse duration shown by the vertical error bars. Horizontal error bars indicate spot size uncertainty, as measured by our pinhole camera and, at low laser energy (about 0.5 mJ), by optical techniques. For the same laser intensity, the gold x-ray pulse duration was found to have a significantly shorter pulse duration, as seen in Fig. 4, and was streak-camera limited at the lowest intensity. This is presumably due to the higher electron densities that one would expect in a high-Z material such as gold, which lead to faster cooling rates.

For the highest laser intensities on target, the conversion efficiency reached about 0.1% (up to 1 mJ) for the x-ray photons with $h\nu \geq 1$ keV. The conversion efficiency increases toward longer wavelengths, implying even higher conversion efficiencies, and therefore even higher brightnesses, in the soft x-ray region. Also, a measured increase in x-ray yield (normalized to the emission volume), with increasing laser intensity, implied a corresponding increase in electron temperature, as expected from previous work [12]. Conversion efficiency for the gold target was found to be 4 times higher than for the aluminum target, due to the higher density of emission lines in this spectral region.

In the most general terms, the duration of emission for a particular x-ray line from a plasma ion is determined

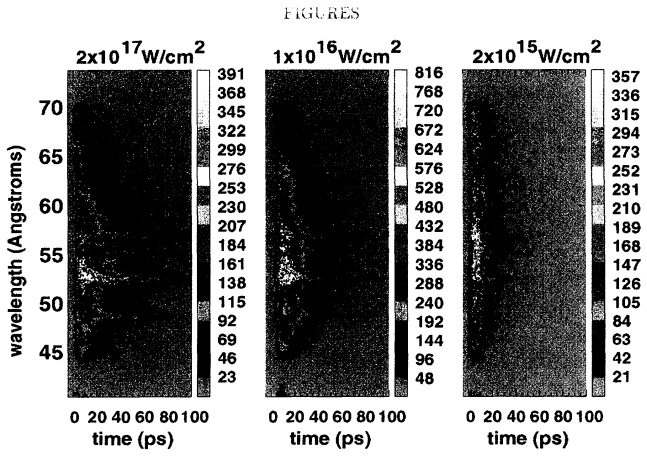

FIG. 1(color). Temporally and spectrally resolved aluminum x-ray emissions obtained for three different laser intensities.

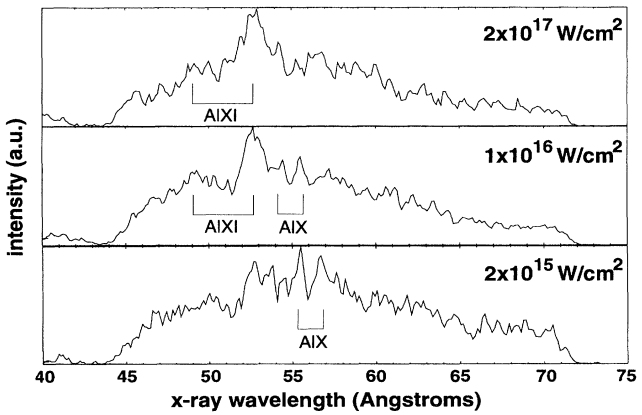

FIG. 2. Aluminum spectra at the peak of emission for three laser intensities showing both the change in peak temperature with laser intensity and the effects of high-density broadening. Spectral resolution is approximately 0.2 Å as determined by long pulse (nanonsecond) illumination of a solid target.

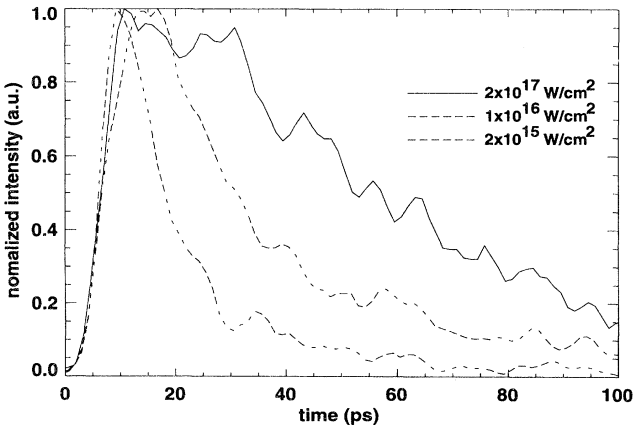

FIG. 3. Comparison of amplitude-normalized temporal profiles of the Al XI $1s^2 2p$-$1s^2 3d$ transition obtained experimentally, showing the reduction of pulse duration with decreasing laser intensity.

by the dynamics of the electron energy-level populations. At high plasma density, these factors depend on the rate of collisions between free electrons and ions. The rise time of the x-ray pulse is roughly determined by the plasma heating time. In the absence of radiative heating, a given region of the plasma is heated either by direct deposition of laser energy or by diffusion of heat from neighboring regions. The decay of the x-ray emission will be controlled by cooling processes, which include both expansion into the vacuum and heat conduction into the colder regions of the solid. For a particular ion, bound-bound emission from any local region of the plasma, at any given time, is determined by the population densities of the various allowed energy levels. More precisely, the emission will be controlled by the source function, defined as the emissivity divided by the opacity, ϵ/κ [13]. For bound-bound emission, the temporal behavior of the source function depends almost linearly on n_u/n_l [14], where n_u is the upper state population density of the transition considered and n_l is the lower state population density. The time history of the emission is then determined by the time history of the upper- and lower-state density ratios.

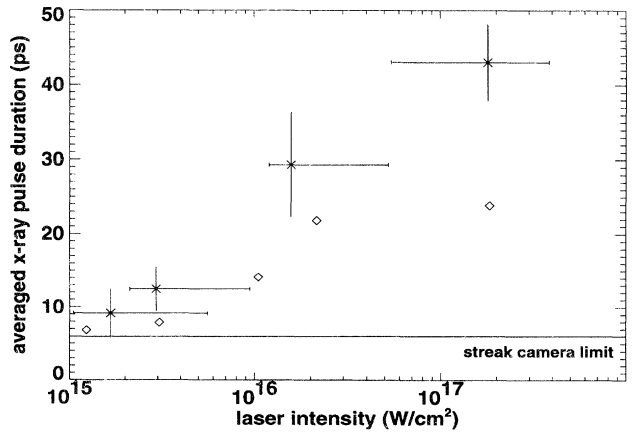

FIG. 4. Duration of the x-ray emission (at $\lambda = 52.4$ Å) versus laser intensity as measured experimentally: aluminum (crosses) Al XI $2p$-$3d$, gold (diamonds).

We can consider an atomic system consisting of n_u and n_l and a free electron population. For the conditions of our experiments with aluminum, lithiumlike ions at high density (which gives rise to continuum lowering), this system is not unreasonable. In a collisionally dominated plasma, the populations of the two bound levels are controlled both by the collisional excitation and collisional deexcitation rates between the two levels and by the collisional ionization and recombination rates between the bound and free states. Because the collisional deexcitation rate per ion is proportional to $n_e/\sqrt{T_e}$, and the collisional excitation rate per ion is proportional to $n_e \exp(-h\nu_{ul}/kT_e)/\sqrt{T_e}$ [15], the ratio of these two rates is controlled by the factor $\exp(-h\nu_{ul}/kT_e)$. For the recombination and collisional ionization rates, the net recombination into any particular level scales as $\exp(\chi/kT_e)$ [16], where χ is the ionization potential. The net rate of change of population between the two bound levels, due to recombination of free electrons, again scales as $\exp(-h\nu_{ul}/kT_e)$. We see that the higher the electron temperature, the smaller the difference between the rates and, therefore, the smaller the change in the ratio n_u/n_l for a given change in the electron temperature. A higher peak electron temperature would mean a slower decrease in emission, which changes linearly with the population ratio, and, therefore, a longer pulse duration for a given cooling rate. As the electron temperature decreases, the difference between the rates begins to change more rapidly. Obviously, if the collision rates are lower than the spontaneous rates there will be very little dependence of pulse duration on electron temperature.

The experimental data clearly show the predicted drop in x-ray pulse duration with decreasing laser intensity. As noted earlier in Fig. 3, the slope of the decay of the x-ray emission is steeper in the lower intensity case, with a corresponding decrease in electron temperature (as seen in Fig. 2). This is consistent with the temporal dependence of x-ray emission on temperature discussed above. Both the Stark broadened emission profiles, shown in Fig. 2, and the intensity dependence of the x-ray pulse duration support the conclusion that the x-ray emission is, in fact, generated in highly collisional regions of the plasma.

In order to maximize the total x-ray emission and minimize the pulse duration, the total number of x-ray emitters should be maximized, while optimizing the temperature. If the peak electron temperature is kept too low, the x-ray intensity will be greatly reduced because the proportion of ions responsible for emission,

in a particular spectral region, will be smaller [17]. The x-ray emission can be kept high by maximizing both the plasma density and the radial dimensions of the region over which the optimal temperature conditions exist. The former means that the shortest laser pulse should be used in order to increase the direct deposition of laser energy at solid density. The latter implies that the laser spot size should be maximized, while keeping the peak laser intensity equal to its optimal value for the laser energy available.

The factors involved in controlling the x-ray pulse duration, described above, involve plasma conditions locally, where the x rays originated. However, due to finite plasma size, a detector external to the plasma will integrate the contributions from each local region along the radiation propagation path. Thus absorption, reemission, escape, stimulated emission, and Doppler shifts must also be considered, which, by coupling the radiation from separate regions, makes the problem a global one. The temporal and spectral characteristics of the observed x-ray emission are determined by the complex interplay between these global effects and local plasma conditions. Since these effects are both time and space dependent, the complexity of the problem is well suited to detailed numerical analysis. As will be discussed in a longer paper, the results of modeling the Al XI $2p$-$3d$ x-ray pulse duration with a one-dimensional hydrodynamics code [18], coupled to a time-dependent detailed-configuration atomic physics package, FLY [19], are in qualitative agreement with the trends that are both predicted by our simple collisional model and observed experimentally in this study. The experimental results reported in this Letter will be useful both for experimentalists, and for further comparison with other detailed numerical models.

This work was partially funded by the National Science Foundation Center for Ultrafast Optical Science, Contract No. PHY8920108. The authors would like to thank G. Mourou for useful discussions.

[1] G. Mourou and D. Umstadter, Phys. Fluids B **4**, 2315 (1992).

[2] M. M. Murnane, H. C. Kapteyn, R. W. Falcone, in *Short Wavelength Coherent Radiation: Generation and Application,* edited by R. W. Falcone and J. Kirz (Optical Society of America, Washington, DC, 1989), Vol. 2, p. 189.

[3] *OSA Proceedings on Short Wavelength Coherent Radiation: Generation and Applications, 1991,* edited by P. Bucksbaum and N. Ceglio (Optical Society of America, Washington, DC, 1991).

[4] S. E. Harris and J. D. Kmetec, Phys. Rev. Lett. **61**, 62 (1988).

[5] D. Umstadter *et al.,* Bull. Am. Phys. Soc. **34**, 1364 (1989); H. Milchberg, I. Lyubomirsky, and C. G. Durfee, III, Phys. Rev. Lett. **67**, 2654 (1991).

[6] M. M. Murnane, H. C. Kapteyn, and R. W. Falcone, Phys. Rev. Lett. **62**, 155 (1989).

[7] D. G. Stearns, O. L. Landen, E. M. Campbell, and J. H. Scofield, Phys. Rev. A **37**, 1684 (1988).

[8] C. Wulker, W. Theobald, F. P. Schafer, and J. S. Bakos, Phys. Rev. E **50**, 4920 (1994).

[9] J. C. Kieffer *et al.,* Appl. Opt. **32**, 4247 (1993).

[10] B. L. Henke *et al.,* J. Opt. Soc. Am. B **3**, No. 11, 1540 (1986).

[11] Corrections for the spectral response of the grating and K edge due to carbon have not been included in the plot.

[12] H. Milchberg and R. Freeman, Phys. Fluids B **2**, 1395 (1990).

[13] D. Mihalas, *Stellar Atmospheres* (W. H. Freeman and Company, San Francisco, 1978).

[14] As n_u/n_l approaches 1, the dependence is only slightly nonlinear.

[15] H. Van Regemorter, Astrophys. J. **136**, 906 (1962); D. M. Cochrane and R. W. P. McWhirter, Phys. Scr. **28**, 25 (1983).

[16] R. H. Huddlestone and S. L. Leonard, *Plasma Diagnostic Techniques* (Academic Press, New York, 1965).

[17] The proportion of ions responsible for emission in a particular spectral region also will be reduced if the temperature is too high.

[18] X. Liu, Ph.D thesis, University of Michigan, 1994.

[19] R. W. Lee *et al.,* J. Quantum. Spectrosc. Radiat. Transfer **32**, 91 (1984).

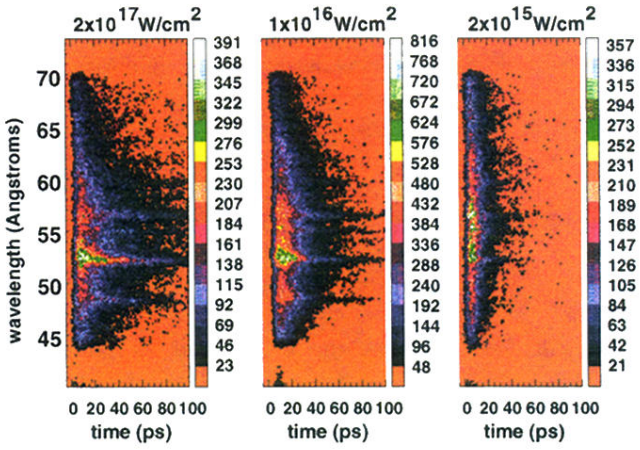

FIG. 1(color). Temporally and spectrally resolved aluminum x-ray emissions obtained for three different laser intensities.