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Enhancement of keV x-ray emission in laser-produced plasmas by a weak prepulse laser

R. Kodama, T. Mochizuki, K. A. Tanaka, and C. Yamanaka
Institute of Laser Engineering, Osaka University, 2-6 Yamada-oka, Suita, Osaka 565, Japan

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X-ray conversion efficiency in a 0.53-μm laser-produced copper plasma is studied in the 1.5–5.0 keV range as a function of laser pulse duration \((\tau_L)\) with a laser intensity of \(1 \times 10^{14}\) W/cm². The efficiency increases as \(\tau_L^{1.3}\) at pulse lengths of less than 400 ps. For a 200-ps pulse duration, an enhancement of the conversion efficiency is observed with the use of a prepulse. The efficiency is found to be proportional to the scale length of a preformed plasma. Enhancement of a factor 3 is observed for the shots with a prepulse.

Laser-produced plasmas have attracted strong interest for its potential use as an x-ray source with their high brightness and small source size. The development of a high-repetitive solid state laser is now making its application to an x-ray lithography source feasible. One of the most important factors for such use is the x-ray conversion efficiency within the range of 1–3 keV photons which are most favorable for x-ray lithography.¹

High emissivity of keV x rays is only achieved in high-temperature, say more than 400 eV, plasmas having sufficient charge states and excitations which are produced under the high intensity irradiation by laser light.² Such high-temperature plasmas can be formed near the underdense region so that opacity against keV photons is generally low. Therefore, the x-ray conversion efficiency will increase in a plasma with a long density scale length.³ There are a few previous studies which indicate that a larger plasma volume increases x-ray conversion efficiencies.⁴⁻⁵ Most of the work to date, however, has not contained a systematic study of the controlled volume dependence of soft x-ray emission.

In this letter we present and discuss the enhancement of x-ray conversion efficiencies in 0.53-μm laser-produced plasmas with variable plasma scale length using Cu planar targets. Two sets of experiments were conducted; the conversion efficiency was measured (A) by varying the laser pulse duration and (B) by introducing a prepulse. In the (A) experiment, the conversion efficiency increases with the pulse duration for pulse widths less than 400 ps and saturates at 1 ns. In (B) we obtained an enhancement of the conversion efficiency by a factor of 3 and found this to be proportional to the scale length of a preformed plasma determined from hydrodynamic calculations and x-ray pinhole images.

Experiments were carried out using two beams of the GEKKO IV Nd:glass laser system. In the (A) experiment, a temporally Gaussian 0.53-μm laser light pulse of variable duration (130 ps–1 ns full width at half-maximum [FWHM]) with no prepulse and in (B) a 0.53-μm main laser pulse of 200 ps (FWHM) preceded by a 1.05-μm prepulse (200 ps) were used. The main and prepulses were focused onto Cu \((Z = 29)\) targets through aspherical lenses of \(f/1.6\) and \(f/8\) with incidence angles of 35° and 22.5° to the target normal, respectively. The intensity of the main laser pulse was fixed at \(1 \times 10^{14}\) W/cm² in the spot diameter of \(250 \times 300 \mu m\) in both modes \((250 \times 300 \mu m)\) represents the minor and major axes of the elliptical shape of the focal spot. In (B) the prepulse laser intensity and the time interval between the two pulses were varied from \(0.2 \times 10^{13}\) to \(9 \times 10^{13}\) W/cm² and from 1 to 3.1 ns, respectively. The prepulse laser spot was the same as the spot of the main laser pulse.

X-ray intensity measurements were performed by using an Al photocathode biplanar x-ray diode (XRDi) with a 25-μm-thick beryllium filter which provided a major spectral sensitivity in the photon energy range from 1.5 to 5.0 keV. The smaller sensitivity is down to an ~1-keV photon range which corresponds to the L spectra of copper. The XRDi was mounted at 22.5° with respect to the target normal. The XRDi used in this experiment is described in Ref. 3. The overall time response of detection was 350 ps so that x-ray signal pulses corresponding to each laser pulse were easily discriminated in time since the time interval between the pre- and main pulses was at least 1.1 ns. The x-ray conversion efficiency is defined as the ratio of the measured x-ray fluence to the main laser energy per unit solid angle. One can obtain the total conversion efficiency by integrating with the angular distribution function over the solid angle. The angular distribution is well approximated to be semispherical for keV x rays.

Images of the x-ray emission above the 1.3-keV range were recorded on KODAK 2494 RAR film using an x-ray pinhole camera (XPHC) with a 31-μm-thick beryllium filter and a pinhole of 10 μm diameter. The XPHC viewed the time-integrated image from the right angle to the target normal.

Figure 1 shows the result from the (A) experiment: the x-ray conversion efficiency \(\eta_x\) as a function of a Gaussian laser pulse duration \(\tau_L\) for a constant laser intensity of \(1 \times 10^{14}\) W/cm² with no prepulse. One can see that the longer the pulse duration the higher the efficiency. For a 400-ps pulse the conversion efficiency is 0.8%/sr which is about 2.5 times larger than that for a 130-ps pulse. For pulses less than 400 ps, the conversion efficiency scales as \(\eta_x \propto \tau_L^{-0.3}\). The observed increase of the conversion efficiency may be attributed to the increased emitting volume, simply because this volume is a function of the laser pulse duration. Detailed analysis of the emitting volume is given in the following paragraph. A longer pulse duration might increase the inverse bremsstrahlung absorption,⁶ which may also be another reason for the increased conversion efficiency. The absorption for an 80-ps pulse, however, could reach as high as
Thus the increased absorption is not enough to explain the above result.

The x-ray emissivity per unit volume (W/cm^3) is a function of the electron temperature and the ion number density. The electron temperature (T_e) increases with pulse duration (r_L) increases. According to the self-regulating model^9 and assuming \( \langle Z \rangle \sim \frac{2}{3}(A T_e)^{1/3} \), it scales as \( T_e \propto r_L^{1/3} \). Here \( \langle Z \rangle \) is the average ionization charge and \( A \) is the atomic number. The x-ray emitting volume is proportional to the density scale length, assuming a planar plasma expansion. The density region responsible for the generation of x rays is assumed to be at critical density. The scale length is inferred to be proportional to \( C_s r_L \), where \( C_s \) is the sound velocity. From these scalings we can estimate that the conversion efficiency varies as \( \eta_e \propto r_L^{29/22} \exp\left(-3.86/r_L^{1/11}\right) \). In this estimation we also assume that the plasma is optically thin and the x-ray pulse duration is approximately equal to the laser pulse duration. This scaling is in reasonable agreement with the experimental results for durations less than 400 ps as shown in Fig. 1. The agreement indicates that the increase in the temperature and the emission volume are most responsible for the increase in the conversion efficiency.

The saturation at the 1-ns pulse duration could be explained by the effect of three-dimensional expansion. Since the three-dimensional expansion can be dominant after \( r_L \sim 800 \text{ ps} \) (\( C_s r_L \sim \text{focal spot diameter} \)), the scale length is limited to the focal spot size, causing the saturation.

Shown in Fig. 2(a) are a typical pinhole image and its axial intensity scan for the single pulse in the (A) experiment and, in Fig. 2(b), the same for the double pulse in the (B) experiment. Although the XPHC images are time integrated and have a finite spatial resolution, these images clearly show that the emitting volume is extended by the prepulse laser. The intensity of the x-ray emission from the prepulse itself was found to be negligible in Fig. 2 as well as in the XRD signal measurement. The first x-ray pulse due to the prepulse was found to be as small as 13% of that from the main pulse even at the maximum prepulse intensity.

Figure 3(a) shows the x-ray conversion efficiency as a function of the prepulse laser intensity at variable pulse delays of 1.1–3.1 ns. It is readily seen in Fig. 3(a) that the conversion efficiency increases with prepulse laser intensity and/or pulse delay. For a 1.8-ns delay, the conversion efficiency is 0.85%/sr at the prepulse laser intensity of \( 1.2 \times 10^{13} \text{ W/cm}^2 \). This efficiency is about three times larger than that without a prepulse. Such enhancement of the conversion efficiency cannot be fully explained by the absorption increase because of the same reason given in the single pulse (A) experiment.

The x-ray conversion efficiency as a function of the preformed plasma scale length is shown in Fig. 3(b). Here the plasma scale length (\( L \)) at the turning point is evaluated from the hydrodynamic calculation taking account of spherical expansion in the XPHC images. The plasma extension at the peak of the main pulse evaluated from the numerical calculations agrees well with that from the XPHC images at the various prepulse conditions. Thus the numerical calculation results can be used for guide lines of the scale length.

The experimental result of the conversion efficiency is found to scale as \( \eta_e \propto L \) (the emission volume) up to the plasma scale length of about 50 \( \mu \text{m} \). The XPHC images also show the increase in the emission volumes as already shown in Fig. 2. As mentioned earlier in the (A) experiment, the electron temperature is also essential to determine the keV x-ray conversion efficiency. However, from the numerical calculation the electron temperature at the peak of the main pulse is not influenced by the plasma scale lengths of up to 50 \( \mu \text{m} \), which are created by the prepulse. On the other hand, the laser absorption length is estimated to be 30–60\( \mu \text{m} \) at the laser temperature \( T_e = 0.6–1 \text{ keV} \). At scale lengths less...
FIG. 3. X-ray conversion efficiencies as a function of (a) a prepulse laser intensity and (b) a plasma scale length for variable delays (△: 1.1 ns, ○: 1.3 ns, ●: 1.8 ns, △: 3.1 ns).

than the limit of the absorption length, the preformed plasma is heated sufficiently by the main laser. Thus the conversion efficiency increases linearly with the scale length or the emission volume.

However, the saturation of the conversion efficiency is found even at scale lengths less than the absorption length for certain prepulse conditions. The saturation begins with the prepulse laser intensity of about 4 × 10^{13} W/cm^2 for the pulse delay of 1.1 ns from Fig. 3(a). From the numerical calculation the electron temperature at the time of the main pulse increases with the prepulse laser intensity for the short time pulse delay such as 1.1 ns. When the electron temperature is so high that the population of L-shell electrons decreases in the preformed plasma, the L-spectra conversion efficiency will saturate at the time of the main heating pulse.

There are some other possible mechanisms on the saturation of the conversion efficiency: the Brillouin backscattering and the condition of geometrical matching between the expanding volume of the preformed plasma and the focusing cone of the main pulse laser. More detailed study is needed for the saturation mechanisms.

In conclusion, we have obtained the dependence of the x-ray conversion efficiency on the laser pulse duration in the 1.5-5.0 keV range from Cu targets irradiated by a 0.53-μm single-pulse laser at 1 × 10^{14} W/cm². The longer pulse duration shows a higher x-ray conversion efficiency, indicating that the emission volume and the electron temperature are essential. For the shots with a prepulse, we obtained the enhancement of the conversion efficiency as much as three times those without prepulse. The conversion efficiency has been found to increase with the preformed plasma scale length as \( \eta_x \propto L \) at a weak prepulse intensity. These results suggest that the conversion efficiency can be significantly improved by optimizing the pulse shape of the laser, for example, by using a tailored laser pulse.

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13. In this simulation, we used the laser spot diameter as the radius of a spherical target when \( r_s \) is greater than the spot diameter and otherwise we used the value of three times as large as the spot diameter. The hydrodynamic computer code used is described in the literature by M. Murakami and K. Nishihara, ILE Quarterly Progress Report ILE. QPR-83-9 (1983), p. 34.