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Dependence of hard x-ray yield on laser pulse parameters in the wavelength-cubed regime

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Conversion efficiency and electron temperature scaling laws are experimentally studied in the wavelength-cubed (λ^3) regime, where a single-wavelength focus allows low energy pulses incident on a Mo target to produce x rays with excellent efficiency and improved spatial coherence. Focused intensity is varied from 2×10^{16} to 2×10^{18} W/cm². Conversion efficiency and electron temperature are best described by a power law for *energy* scaling while an exponential law best describes the scaling of these parameters with *pulse duration*. © 2004 American Institute of Physics.

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X-ray generation from an ultrafast-laser-produced plasma is increasingly obtaining the attention of scientists in a diversity of fields. Because this kind of x-ray source has its own unique features—high efficiency and short pulse duration—it has been used in many experiments, such as imaging, time-resolved diffraction, spectroscopy and microscopy of transient physical, chemical, or biological phenomena. With the added concentration of light to a very small spot, minimal x-ray source size can be obtained. This offers the advantages of high brightness and good spatial coherence. Thus more demanding applications can be addressed.

For any kind of application, x-ray yield is one of the most important issues. On the one hand, several groups have increased x-ray yield by modifying the target surfaces irradiated by laser pulses. 1-4 On the other hand, some groups have improved the x-ray yield by manipulating laser pulse characteristics. For example, p-polarized large oblique-incidence yields x rays much more efficiently than s-polarized, smallincident-angle illumination.⁵ By varying the intensity of prepulses and their time delay relative to the main pulse, the density scale length of the plasma can be controlled, and the laser energy absorption and x-ray yield are optimized.^{6,7} Eder et al.8 found that the most intense laser pulse was not necessarily the optimal condition for hard x-ray production. The optimal intensity for $K\alpha$ x-ray yield was studied experimentally and theoretically by Ziener et al.7 and Reich et al.9 Definitely, laser parameters, such as pulse energy, pulse duration, focal spot size, focal intensity, polarization, incidence angle, and the intensity contrast ratio (of main-pulse to prepulse and/or amplified spontaneous emission), are important factors for x-ray yield. Still, more data are needed to determine the optimal conditions of laser-based x-ray generation.

Scaling of x-ray yield with pulse energy has been investigated with targets of Cu, Al, C, SiO₂.5,10-12 A few publications 10,13 have studied scaling with pulse duration in the long pulse range. Here we focus on the relationships of x-ray conversion efficiency with pulse energy and pulse duration in the tightly focused regime (spot size $\sim \lambda^2$, where λ is laser wavelength). We define the x-ray conversion efficiency, η , as the ratio of x-ray pulse energy emitted in 2π sr in front of a target to the laser pulse energy. Recently the spectroscopy of x rays from metallic targets generated in the "relativistic wavelength-cubed (λ^3) " regime was experimentally studied in our lab.¹⁴ We tightly focused severalcycle pulses to a near-single-wavelength spot by a combination of a deformable mirror with an f/1 paraboliodal mirror, reaching relativistic intensity $(2 \times 10^{18} \text{ W/cm}^2)$. We dubbed this the "relativistic λ^3 " regime because the volume of the focused pulse is $<(3\lambda)^3$. This, in turn, generates a very brief, high intensity hard x-ray burst with a minimum dimension less than 5 μ m, ¹⁴ dramatically improving spatial coherence which scales as the inverse square of spot size for such sources. In the current work, we extend the study of the energy scaling law (at a fixed pulse duration of 22 fs) into λ^3 regime. The pulse duration scaling is systematically investigated to the shorter limit of pulse duration (from 570 down to 22 fs) for a constant laser pulse energy of 1.1 mJ, while maintaining a λ^2 focus. Molybdenum (Mo) was chosen as a target material in our experiments. The photon energy of its K-line emissions is \sim 17.5 keV, which is suitable for mammography. 15

These experiments utilize a compact table-top Ti:sapphire chirped-pulse-amplified laser system. This laser produces a train of pulses with 1.1 mJ pulse energy, 22 fs pulse duration, at 400 Hz repetition rate. In the experiments, half

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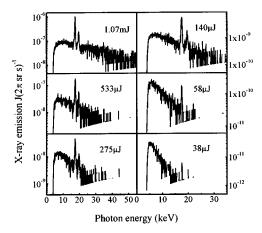


FIG. 1. X-ray spectra of a Mo target. The laser pulse duration is fixed at 22 fs, but the pulse energy is varied from 23 μ J to 1.07 mJ by a waveplate.

of the laser pulse energy arrives on target. The p-polarized pulses are focused onto the Mo target at a 45° incidence angle with a spot size of $1.2 \times 1.4 \mu \text{m}^2$ (full width at half maximum). In our experiments, a combination of a waveplate and polarizer is used to vary the pulse energy, while the pulse duration is varied by changing the separation between compressor gratings. X rays are generated from a thick Mo disk that is rotated and translated to show a fresh area to each shot. An x-ray detector (XR-100CZT/, Amptek, Inc.) is installed at 40° to the target normal (opposite the incident beam and in the incidence plane), 55 cm from the plasma, to measure the x-ray spectrum. A pinhole with a proper diameter is placed 5.5 cm in front of the detector to avoid the pileup effect on the measured spectrum. A pair of magnets is positioned between the plasma source and the beryllium (Be) chamber window to prevent x-ray fluorescence from being produced on the window by high-energy electrons. The spectra were accumulated over $\sim 10^5$ laser shots.

One experiment is to study the scaling of x-ray conversion efficiency with laser pulse energy, E. The series of spectra shown in Fig. 1 are obtained by varying the laser pulse energy from 23 µJ to 1.07 mJ, with the pulse duration fixed at 22 fs. It is clear that $K\alpha$ and $K\beta$ line emissions are sitting on a bremsstrahlung continuum background. The total x-ray energy and the ratio of line-to-background emission energies are both decreased with diminishing laser-pulse energy. In the 58 μ J data, the $K\beta$ peak falls below the detection limit. When the pulse energy is reduced to 38 μ J, both line peaks almost vanish. With reduced pulse energy the magnitude of the bremsstrahlung background slope becomes larger, indicating a diminishing electron temperature, T_e , as shown in Fig. 2(a). From the spectra in Fig. 1, we calculate η_{α} , $\eta_{\alpha\beta}$, and η_{tot} (the conversion efficiencies of, $K\alpha$ line emission, $K\alpha$ plus $K\beta$ line emissions, and total x-ray emission between 4 and 55 keV, respectively) for different laser pulse energies. Also, T_e can be calculated from the slope of bremsstrahlung background (in our detection window, we find a single exponential decay of x-ray spectral power with photon energy). After carefully analyzing the data obtained in our experiments, all of the efficiencies obey the power law η $\propto E^{\gamma}$. The fitted results are shown by solid lines in Fig. 2(a). For η_{tot} , we find $\gamma_{\text{tot}} \approx 1.59 \pm 0.15$, and for η_{α} and $\eta_{\alpha\beta}$, we find $\gamma_{\alpha} \approx 1.50 \pm 0.16$ and $\gamma_{\alpha\beta} \approx 1.47 \pm 0.15$, respectively.

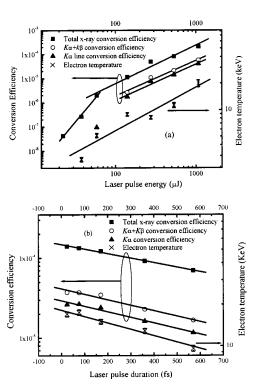


FIG. 2. Conversion efficiencies and electron temperature (a) as functions of laser pulse energy for τ =22 fs, plotted on a log-log scale, and (b) as functions of laser pulse duration for E=1.07 mJ, plotted on a linear-log scale, respectively. Data points represent the values calculated from the experimental data. Solid curves are fitted results from the models. The error bars of T_e come from the fitting errors of the bremsstrahlung continua slope. Left axis represents the conversion efficiency, and right axis electron temperature

Here, T_e is also modeled by a power law, and $\gamma_{T_e}{\approx}0.51{\pm}0.08$, which is in agreement with theoretical work. 9,16 The similarity of the power laws for temperature and for efficiency can be explained by the fact that suprathermal electrons are responsible for x-ray production. With respect to the balance of x rays generated in total emission and line emissions: the curve for the total x-ray efficiency (η_{tot}) goes up slightly faster than the curves for η_{α} and $\eta_{\alpha\beta}$, i.e., the ratio $\eta_{\rm tot}/\eta_{\rm line}$ increases with the pulse energy. This means, in our situation, the bremsstrahlung emission increases slightly faster than line emissions with laser pulse energy. In Andreev's work, 12 with 700 fs, 248 nm laser pulses interacting with carbon, the experiment also yielded $\gamma_{\text{tot}} > \gamma_{\text{line}}$. In the low pulse energy range up to 100 μ J [three left-most squares in Fig. 2(a)], η_{tot} increases faster with the pulse energy having a scaling exponent of \sim 4. This deviation begins as the electron temperature drops. We interpret this to be caused by the fact that the peak of the x-ray energy distribution is shifting toward or out of the lower limit of our detection window as laser pulse energy decreases. Also, the line emission efficiency indicated by the leftmost triangular point at 58 μ J is significantly below the extrapolated value. This is because the peak of the electron temperature distribution has moved too far below K-shell energy.

In the second experiment, we study the scaling of x-ray conversion efficiency with laser pulse *duration*, τ , which is varied from 22 to 570 fs for fixed laser pulse energy of 1.1 mJ. The x-ray spectra (not shown) are similar to those obtained in the previous case. As pulse duration is increased,

the intensity of the K-shell line emissions diminishes with respect to the background, but is always present within the investigated range. Similarly, the slope of the background emission increases with pulse duration. This means T_e decreases for longer pulse durations or lower intensity. The calculated η_{tot} , η_{α} , $\eta_{\alpha\beta}$, and T_e at different pulse durations are shown in Fig. 2(b). A power law, however, cannot model the data in Fig. 2(b). Schnürer et al. 10 found that, for pulses longer than 120 fs, the x-ray yield was described by I_x $\propto I_{\text{laser}}^{1.7} (I_{laser} \propto \tau^{-1})$ and saturation of the x-ray emission appeared for pulse durations shorter than 120 fs. His model failed to describe the saturation. We also find an onset of saturation in our experiment. The reason for this is x-ray reabsorption in the bulk target. More-energetic electrons produce x-ray photons deeper inside the target because of their higher energy and smaller collision cross-section. So, reabsorprion is stronger for these photons. A reduction in $K\alpha$ yield at laser intensities similar to ours was previously discussed by Eder et al.8 and Ewald et al.17

Here we find empirically, that a single exponential decay model, $\eta \propto \exp(-\tau/\beta)$, fits our data well. The fitted results are shown by solid lines in Fig. 2(b), where exponents are $\beta_{\text{tot}} = 0.78 \pm 0.04 \text{ ps}, \ \beta_{\alpha} = 0.65 \pm 0.08 \text{ ps}, \ \beta_{\alpha\beta} = 0.67 \pm 0.09 \text{ ps},$ and $\beta_{T_e} = 0.95 \pm 0.11$ ps. Again, both η and T_e obey the same law. In this experiment, T_e decreases more slowly than η as laser intensity decreases, which is similar to the results of the first experiment. Because $\beta_{tot} > \beta_{\alpha}$ and $\beta_{\alpha\beta}$, the bremsstrahlung emission decreases slightly slower than line emissions with reduced intensity, i.e., increased pulse duration. This is opposite to the first experiment, where the bremsstrahlung emission decreases slightly faster than line emissions as intensity is decreased. Chichkov et al. 13 inferred a simple x-ray scaling law with laser pulse energy and duration. Unfortunately, it does not describe our data. Thus, the mechanism of x-ray generation is more complicated than that represented by his model, at least in the tightly focused regime.

Two-dimensional particle-in-cell simulations with fewcycle relativistic laser pulses focused to a single-wavelength spot size onto a short density scale length $(L/\lambda = 3-5)$ plasma slabs studied in detail plasma heating and particle acceleration.¹⁸ Four different groups of electrons were identified in the phase space distribution as contributing to the electron acceleration. One group contains electrons that are pushed by the laser pulse similar to a snowplow and which are concentrated at the front side of the pulse forming a solitary electrostatic wave with up to four times critical density. Another group contains electrons with the highest energy, created behind the laser pulse. They are accelerated by the electrostatic field that is set up by ponderomotive evacuation of the electrons within the laser pulse. While the laser pulse is reflected from the plasma, the high-energy particles continue to move forward inertially into the bulk material. Fast particles penetrating into the solid are responsible for x-ray production. A shorter pulse duration and a smaller spot size generate a higher laser intensity for a given pulse energy which increases the conversion efficiency in the λ^3 regime. Tight focusing to a wavelength spot might also to help to avoid filamentation, beam breakup, and the growth of plasma instabilities.¹⁹

In conclusion, scaling laws, for both η and T_e , are best described by a power law, $\propto E^{\gamma}$, for energy scaling and an exponential law, $\propto \exp(-\tau/\beta)$, for pulse-duration scaling. With a 22 fs pulse duration, we obtain $\gamma_{\eta} \approx 1.5$ for efficiencies, and $\gamma_{T_e} \approx 0.5$ for electron temperature, respectively. At a pulse energy of 1 mJ, we obtain $\beta_{\eta} \approx 0.72$ ps for efficiencies, and $\beta_{T_e} \approx 0.95$ ps for electron temperature. Our model will be important for optimizing parameters to obtain good x-ray yield from laser-based x-ray sources. These energy and pulse duration scaling laws, measured in the tightly focused regime, are not consistent with commonly used single-parameter (intensity only) scaling laws for either x-ray efficiency or electron temperature.

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- ¹S. P. Gordon, T. Donnelly, A. Sullivan, H. Hamster, and R. W. Falcone, Opt. Lett. 19, 484 (1994).
- ²T. Nishikawa, H. Nakano, N. Uesugi, M. Nakao, and H. Masuda, Appl. Phys. Lett. **75**, 4079 (1999).
- ³ Y. Hironaka, Y. Fujimoto, K. Nakamura, K. Kondo, and M. Yoshida, Appl. Phys. Lett. **74**, 1645 (1999).
- ⁴G. Kulcsár, D. AlMawlawi, F. W. Budnik, P. R. Herman, M. Moskovits, L. Zhao, and R. S. Marjoribanks, Phys. Rev. Lett. 84, 5149 (2000).
- ⁵ U. Teubner, J. Bergmann, B. V. Wonterghem, F. P. Schäfer, and R. Sauerbrey, Phys. Rev. Lett. **70**, 794 (1993).
- ⁶S. Bastiani, A. Rousse, J. P. Geindre, P. Audebert, C. Quoix, G. Hamoniaux, A. Antonetti, and J.-C. Gauthier, Phys. Rev. E 56, 7179 (1997).
- ⁷ Ch. Ziener, I. Uschmann, G. Stobrawa, Ch. Reich, P. Gibbon, T. Feurer, A. Morak, S. Düsterer, H. Schwoerer, E. Försyer, and R. Sauerbrey, Phys. Rev. E 65, 066411 (2002).
- ⁸D. C. Eder, G. Pretzler, E. Fill, K. Eidmann, and A. Saemann, Appl. Phys. B: Lasers Opt. 70, 211 (2000).
- ⁹Ch. Reich, P. Gibbon, I. Uschmann, and E. Försyer, Phys. Rev. Lett. 84, 4846 (2000).
- ¹⁰ M. Schnürer, R. Nolte, A. Rousse, G. Grillon, G. Cheriaux, M. P. Kalachnikov, P. V. Nickles, and W. Sandner, Phys. Rev. E 61, 4394 (2000).
- ¹¹ J. D. Kmetec, C. L. Gordon III, J. J. Macklin, B. E. Lemoff, G. S. Brown, and S. E. Harris, Phys. Rev. Lett. 68, 1527 (1992).
- ¹² A. A. Andreev, U. Teubner, I. V. Kurnin, and E. Föster, Appl. Phys. B: Lasers Opt. 70, 505 (2000).
- ¹³ B. N. Chichkov, C. Momma, A. Tünnermann, S. Meyer, T. Menzel, and B. Wellegehausen, Appl. Phys. Lett. 68, 2804 (1996).
- ¹⁴B. Hou, J. Nees, M. Wilcox, and G. Mourou. Conference on Lasers and Electro-Optics, 1–6 June, 2003, Baltimore, MD, CMZ3.
- ¹⁵ A. Krol, A. Ikhlef, J. C. Kieffer, D. A. Bassano, C. C. Chamberlain, Z. Jiang, H. Pépin, and S. C. Prasad, Med. Phys. 24, 725 (1997).
- ¹⁶P. Gibbon and A. R. Bell, Phys. Rev. Lett. **68**, 1535 (1992).
- ¹⁷F. Ewald, H. Schwoerer, and R. Sauerbrey, Europhys. Lett. **60**, 710 (2002).
- ¹⁸G. I. Dudnikova, V. Yu. Bychenkov, A. Maksimchuk, G. Mourou, J. Nees, S. G. Bochkarev, and V. A. Vshivkov, Phys. Rev. E 67, 026416 (2003).
- ¹⁹G. Mourou, Z. Chang, A. Maksimchuk, J. Nees, S. V. Bulanov, V. Yu. Bychenkov, T. Zh. Esirkepov, N. M. Naumova, F. Pegoraro, and H. Rohl, Plasma Phys. Rep. 28, 12 (2002).