Generation of picosecond hard-x-ray pulses in a femtosecond-laser-driven x-ray diode

Ulf Hinze, André Egbert, and Boris Chichkov

Laser Zentrum Hannover e.V., Hollerithallee 8, D-30419 Hannover, Germany

Klaus Eidmann

Max Planck Institut für Quantenoptik, Hans Kopfermann Strasse 1, D-85748 Garching, Germany

Received March 30, 2004

The temporal characteristics of ultrashort hard-x-ray pulses generated in a femtosecond-laser-driven x-ray diode are investigated for what is believed to be the first time. Copper K_α x-ray pulses with a duration of a few picoseconds are measured with a jitter-free x-ray streak camera. © 2004 Optical Society of America

OCIS codes: 140.7090, 320.2250, 320.5390, 340.7480.

The generation of ultrashort hard-x-ray pulses has become of much interest for investigations of ultrafast phase transitions, lattice dynamics, chemical reactions, etc. A promising means of generating ultrashort hard x rays (in the 1–100-keV energy range) is using low-intensity femtosecond laser pulses in combination with the conventional electron acceleration technique. The femtosecond laser pulses induce photoemission of electrons from a photocathode. The electron bunch is then accelerated toward a high-Z target material by the application of an external high-voltage electric field. This arrangement provides an ultrafast electron gun. Hard x rays are produced by bremsstrahlung of accelerated electrons and characteristic emission inside the target. Our work extends the results of Chen *et al.*[1] and more recently of Girardeau-Montaut *et al.*,[2] who developed x-ray diodes driven with nanosecond and picosecond laser pulses.

In contrast with laser-plasma x-ray sources, which require high-power laser systems (see, e.g., Ref. 3 and references therein), in an x-ray diode one can use low-energy, high-repetition-rate, commercially available femtosecond lasers. The generated hard x rays are synchronized with the femtosecond laser pulses, which is important for time-resolved applications. Using a slightly modified setup of the electron gun, one can generate both ultrashort x rays and electron pulses. Since the electron-scattering cross sections are several orders of magnitude larger than those of hard x rays, ultrashort electron pulses are attractive for time-resolved diffraction studies. This possibility was demonstrated by Ihee *et al.*,[4] and recently femtosecond electron diffraction was used for direct measurement of ultrafast atomic motion.[5]

In this Letter we report on our recent progress[6,7] in the temporal characterization of ultrashort x-ray pulses generated in a femtosecond-laser-driven x-ray diode. The experimental setup (Fig. 1) consists of three main parts: a femtosecond laser, the x-ray diode, and an x-ray streak camera for the detection and characterization of the x-ray pulses. A 1-kHz Ti:sapphire femtosecond laser with a pulse duration of 150 fs and a laser wavelength of 780 nm is operated at typical pulse energies of 300 μJ. The laser pulses are split into two parts by a beam splitter: 80% are directed to the streak camera's optical trigger input, and the remaining part of the energy is used for the generation of ultrashort electron bunches and hard x rays.

The x-ray diode consists of a vacuum chamber containing two electrodes at a distance of 10 mm. The electrodes are polished copper rods (diameter of 10 mm) cut at an angle of 45°. The anode is connected to a high-voltage power supply (0–100 kV), whereas the cathode is set to ground potential.

The laser light is directed by an $f = 22$ cm lens through a quartz window onto the cathode surface. The typical laser spot radius at the cathode surface is 0.25 mm. Each laser pulse generates a bunch of electrons that are accelerated toward the anode (copper target). When the electrons hit the target, x rays (bremsstrahlung and characteristic line emission) are produced. The x rays leave the vacuum chamber

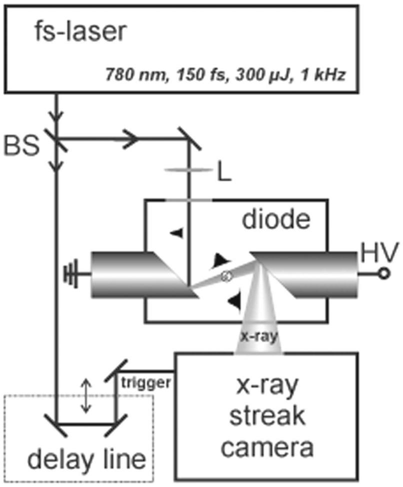

Fig. 1. Experimental setup consisting of a femtosecond laser system, x-ray diode, and streak camera. BS, beam splitter; L, lens; HV, high voltage.

through a 1-mm-thick beryllium window. The number of copper K_α x-ray photons generated at a 30-μJ laser pulse energy and 60-kV acceleration voltage is of the order of 10^9 photons/s, corresponding to 0.5 μJ/s in 2π sr. A detailed characterization of the x-ray diode in terms of spectral properties, conversion efficiency, and x-ray flux can be found in Ref. 7.

For signal analysis an ultrafast x-ray streak camera[8] is used. It accumulates many shots by means of a laser-triggered GaAs switch. For x-ray pulses a temporal resolution of 1.3 ps was calculated.[9] The camera is equipped with a beryllium window (thickness of 1 mm) and a KI photocathode. The distance between the x-ray source and the entrance slit of the streak camera is 8 cm. The streak images are taken by a CCD camera (1024 × 1024 pixels). The horizontal image axis corresponds to temporal resolution; a single pixel is 300 fs wide. The vertical direction provides spatial resolution. The exposure time of the CCD camera can be varied from milliseconds up to several tens of minutes. The experimental points presented in this Letter were recorded at integration times between 60 and 480 s. Note that the shape of the x-ray signal recorded by the streak camera and the position in time do not change when the integration time is varied from a few hundred milliseconds to 10 min.

Typical CCD images are shown in Fig. 2. These images show a strong vertical line in the center, which is a shadow image of the entrance slit produced by hard-x-ray photons propagating through the streak camera photocathode without being converted into electrons. These x rays directly hit the phosphor screen in front of the CCD camera.

The streak signal, produced by hard x rays converted into electrons, appears in the right half of the CCD images shown in Fig. 2 as a slightly curved trace. The shape of the trace is warped by the off-axis electrons, which are traveling slightly longer pathways in the streak tube. The resulting intensity curve (insets in Fig. 2) is fit by a Gaussian profile. From this profile the FWHM and the x-ray pulse duration are determined.

The measured values of the x-ray pulse duration as a function of the acceleration voltage are shown in Fig. 3 for different laser pulse energies. The corresponding number of electrons per pulse was determined by measuring the diode current and is given in the inset in Fig. 3.

As can be seen, the width of the x-ray pulses measured in our experiments varies from 3 to 21 ps. The x-ray pulse duration becomes shorter when the laser pulse energy (i.e., the number of photoelectrons) is decreased or when the acceleration voltage is increased. In the first case the space-charge broadening effects and in the second case the acceleration time are reduced. Although the number of photoelectrons grows with the accelerating voltage (see Fig. 3), the x-ray pulse duration decreases. For these experimental conditions the reduction of the acceleration time plays a more important role than the space-charge broadening.

In simulations[10] of the experimental data the influence of both space-charge effect Δt_{sp} and initial energy spread Δt_e on hard-x-ray pulse duration $\Delta t_{\mathrm{total}}$ is considered as a Gaussian convolution $\Delta t_{\mathrm{total}}^2 = \Delta t_{\mathrm{sp}}^2 + \Delta t_e^2 + \tau_0^2$ with

$$\Delta t_{\mathrm{sp}} = \frac{e^{1/2} m^{1/2} d^2 N}{\sqrt{2}\,\pi U^{3/2} \epsilon_0 r^2}\,, \qquad \Delta t_e = \frac{\Delta v_0 m d}{eU}\,, \tag{1}$$

where τ_0 is the laser pulse duration, e is the electron charge, m is the electron mass, d is the electrode distance, N is the number of generated electrons per pulse, U is the acceleration voltage, ϵ_0 is the vacuum permittivity, r is the electron-beam radius, and Δv_0 is the electron velocity spread at the photocathode surface.

Since the electron photoemission is intensity dependent, we must take into account that the size of the area in which photoelectron emission occurs (the electron-beam radius) changes with the laser pulse energy. For an estimate of Δt_{sp} we use $r = r_0 \ln(E/E_{\mathrm{th}})$, where $r_0 = 0.25$ mm is the laser spot radius and $E_{\mathrm{th}} = 1$ μJ is the detection threshold energy. Δt_{sp} depends on the number of electrons N per pulse, which is determined experimentally (see Fig. 3). With these data the spread of the electron pulse due to space-charge effect

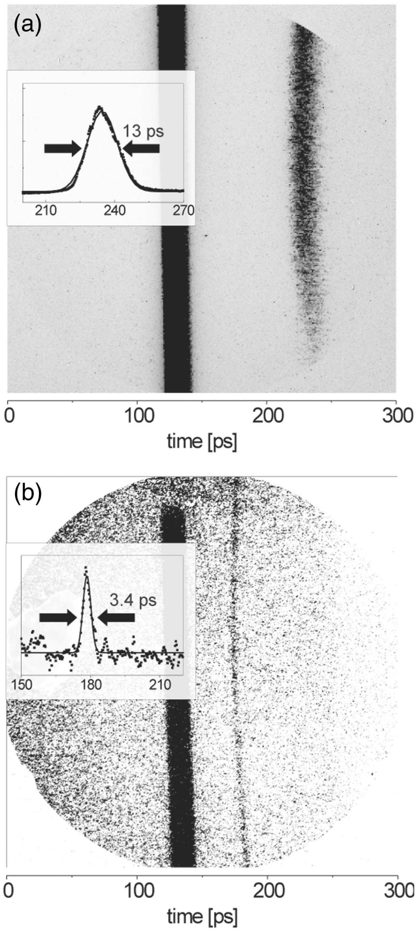

Fig. 2. Typical streak camera images. The traces in the right half of these images are produced by ultrashort copper K_α x-ray pulses recorded at (a) 30 μJ, 40 kV and (b) 3 μJ, 90 kV. Insets, intensity distributions and hard-x-ray pulse durations.

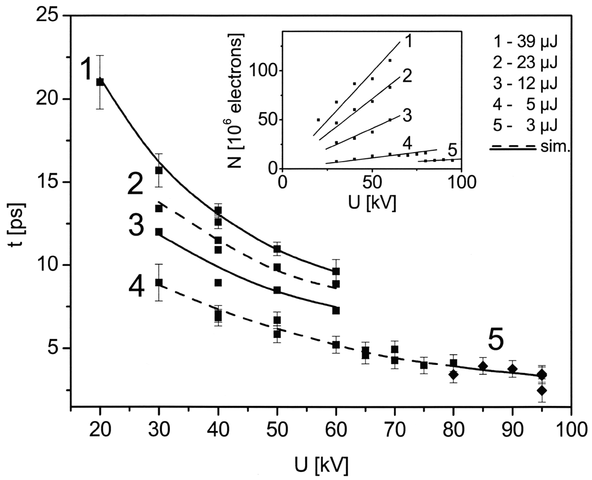

Fig. 3. Dependence of the measured x-ray pulse durations on the acceleration voltage and laser pulse energy. Solid and dashed curves provide calculated values. The inset shows the corresponding number of electrons per pulse.

Δt_{sp} is calculated. The resulting values for Δt_{sp} vary from 21 ps (for 20 kV and 39 μJ) to 3 ps (for 95 kV and 3 μJ).

For an estimate of Δt_e we use available experimental data[11] for initial electron energy spread $\Delta E = 1$ eV of photoelectrons produced by multiphoton emission from a copper target. This energy spread corresponds to electron velocity spread $\Delta v_0 \simeq 5.9 \times 10^5$ m/s. With these data Δt_e is calculated; it varies between 1.7 ps (for 20 kV) and 0.3 ps (for 95 kV).

Thus the temporal broadening of the electron pulses in our experiments is dominated by the space-charge effect. The calculated values of total x-ray pulse duration $\Delta t_{\mathrm{total}}$ are plotted by solid and dashed curves in Fig. 3. These results are in good agreement with the experimental data. The shortest x-ray pulse (3 ps, or 2.7 ps after unfolding with the streak camera's time resolution of 1.3 ps) is produced at an acceleration voltage of 95 kV and a laser pulse energy of 3 μJ. This corresponds to 10^7 electrons and 10^5 hard-x-ray photons per pulse. Note that these measurements are performed close to the detection limit of our streak camera. Reducing the laser pulse energy allows the number of generated electrons to be reduced by a factor of 10. In this case the space-charge broadening [see Eq. (1)] will be reduced by the same factor, and both broadening contributions will become equal, $\Delta t_{\mathrm{sp}} = \Delta t_e = 0.3$ ps. This will result in a hard-x-ray pulse duration of less than 500 fs. Every single hard-x-ray pulse will contain 10^4 photons. With commercially available 250-kHz femtosecond laser systems providing pulse energies up to 4 μJ, it will be possible to generate 500-fs copper K_α x-ray pulses with as much as 2.5×10^9 photons/s. Note that with this laser system we have already measured x-ray photon fluxes[7] exceeding 2×10^{10} photons/s.

In conclusion, to the best of our knowledge, the experiments presented here demonstrate for the first time that 3-ps copper K_α x-ray pulses can be generated in an x-ray diode driven by femtosecond laser pulses. Certainly, these results do not represent any physical or technical limits. By further improvement of the source setup, e.g., by reducing the distance between the electrodes, optimizing their geometry, using the third harmonic of Ti:sapphire laser radiation, one can reduce the x-ray pulse duration to 300–500 fs by still providing more than 10^4 hard-x-ray photons per pulse.

The authors thank A. Assion and T. Baumert for their cooperation and the Deutsche Forschungsgemeinschaft for financial support. U. Hinze's e-mail address is hi@lzh.de.

References

1. P. Chen, I. V. Tomov, and P. Rentzepis, J. Chem. Phys. **104,** 10001 (1996).
2. J. P. Girardeau-Montaut, B. Kiraly, C. Girardeau-Montaut, and H. Leboutet, Nucl. Instrum. Methods Phys. Res. A **452,** 361 (2000).
3. E. Fill, J. Bayerl, and R. Tomassini, Rev. Sci. Instrum. **73,** 2190 (2002).
4. H. Ihee, V. A. Lobashov, U. M. Gomez, B. M. Goodson, R. Srinivasan, C. Y. Ruan, and A. H. Zewail, Science **291,** 458 (2001).
5. J. Cao, Z. Hao, H. Park, C. Tao, D. Kau, and L. Blaszczyk, Appl. Phys. Lett. **83,** 1044 (2003).
6. A. Egbert, B. Mader, B. Tkachenko, C. Fallnich, B. N. Chichkov, H. Stiel, and P. V. Nickles, Appl. Phys. Lett. **81,** 2328 (2002).
7. A. Egbert and B. N. Chichkov, Proc. SPIE **4978,** 92 (2003).
8. C. Belzile, J. C. Kieffer, C. Y. Cote, T. Oksenhaendler, and D. Kaplan, Rev. Sci. Instrum. **73,** 1617 (2002).
9. F. Pisani, U. Andiel, K. Eidmann, K. Witte, I. Uschmann, A. Morak, E. Förster, and R. Sauerbrey, Appl. Phys. Lett. **84,** 2772 (2004).
10. B.-L. Quian and H. E. Elsayed-Ali, J. Appl. Phys. **91,** 462 (2002).
11. M. Aeschlimann, C. A. Schmuttenmaer, H. E. Elsayed-Ali, R. J. D. Miller, C. Cao, Y. Gao, and D. A. Mantell, J. Chem. Phys. **102,** 8606 (1995).