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Convergent-beam diffraction of ultra-short hard X-ray pulses focused by a capillary lens

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ABSTRACT We report the generation of diffraction patterns using X-ray pulses from a fs-laser plasma focused by an X-ray capillary lens to a spot size smaller than 100 μm. A thin moving iron tape is irradiated at 10 Hz with 200 mJ/130 fs titanium–sapphire laser pulses. Fe K_α ($\lambda = 0.194$ nm) radiation emitted from the rear side of the tape is focused to a spot by the capillary lens resulting in an intensity enhancement of about 1600. The ability of this system to effectively allow diffraction from samples of micron size is demonstrated by producing well-illuminated diffractograms from a Si(111) crystal with good signal to noise ratio obtained in only about 10 s. The different path lengths of propagation in the lens induce an angle-encoded delay of the converging X-ray pulse and thus provide the possibility of realizing a novel ultra-fast streak camera.

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1 Introduction

Recent progress in using fs lasers to generate ultra-short intense X-ray pulses has stimulated the development of new diagnostic and imaging techniques in physics, chemistry, biology and material sciences. These include time-resolved X-ray diffractometry [1–5], ultra-fast X-ray absorption [6, 7] and testing of optical components for fourth-generation light sources. Due to the limited number of hard X-ray photons emitted from the laser plasma, most experiments require a substantial number of shots to be accumulated on the detector. Reducing the necessary exposure time can be achieved by locating the target as close as possible to the detector or by focusing the radiation by bent crystal mirrors [8] or elliptical specularly reflecting mirrors [9]. A different efficient way to increase the photon flux on the detector involves using capillary optics [10, 11] for focusing the X-rays [12], e.g. via a converging or a half lens made of numerous capillaries, i.e. a polycapillary X-ray lens. In addition to the intensity enhancement, converging X-rays are advantageous in many cases to determine the crystallographic structure of complex systems [13, 14] including biomolecules. Recently, focusing of fs-laser-generated X-ray pulses by means of a capillary lens has been demonstrated [15]. In that work Cu K_α radiation and radiation from Mo and W from thin wires was focused by a polycapillary lens and the cross correlation of the X-rays and the laser pulses was determined using the (400) reflection from a silicon crystal. The pulse duration of the X-rays after the lens was found to be less than 1 ps. However, in that study typical exposure times range from several minutes up to one hour.

In this paper we demonstrate the usage of an efficient capillary lens in an arrangement that creates a distinctive diffracted image from a micron-size segment of a single-crystal surface within a few seconds, corresponding to a reduced number of laser shots. In contrast to Tomov et al. [15], our method utilizes a more stable and reproducible target setup and short-wavelength production using movable thin metal tapes, capable of producing about $3 \times 10^9 K_\alpha$ photons/Sr per shot [16].

2 Experimental setup

In our setup the polycapillary lens is operated outside the vacuum chamber which houses the tape-drive mechanism (see Fig. 1). This approach greatly simplifies the measurement and alignment procedure of both the X-ray lens and the sample under study. As a result of our specific setup and pump laser characteristics our typical exposure times vary from a single shot to 300 shots, i.e. 30 s at 10-Hz repetition rate. Moreover, this setup utilizes well-defined convergent X-ray beams and may be applied to build a new type of ultra-fast X-ray streak camera.

The Fe K_α radiation was produced, as is well known, by illuminating a solid target with a fs laser beam. The hot electrons generated at the front plasma surface when a high-intensity ultra-short laser beam is incident on a solid target interact with the solid material itself producing bremsstrahlung radiation and line emission. In the case of a mid-Z material, K_α emission is found to dominate the spectrum [17]. The K_α line

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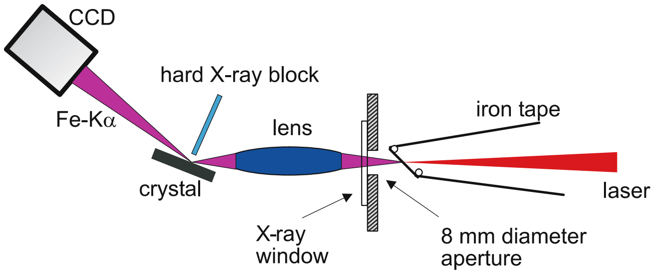

FIGURE 1 Experimental setup used for obtaining convergent-beam diffractograms. 130-fs laser pulses from ATLAS, focused to a spot about 100 μm in diameter, irradiate a slow-moving 20-μm-thick iron tape and generate hot electrons that produce Fe K_α radiation at 6.4 keV. The X-ray pulses are focused by the capillary lens onto a crystal and diffracted to an X-ray CCD. A Bragg line is produced on the CCD along which the time delay varies over 500 fs, thus realizing a new type of ultra-fast X-ray streak camera

can be optimized over a range of illumination conditions. The best scheme for hot-electron generation is when a p-polarized laser pulse is obliquely incident on the target. Another important parameter to adjust is the position of the target with respect to the focal point of the focusing optics [16, 17].

The experimental system is shown in brief in Fig. 1. Laser pulses from the ATLAS titanium–sapphire installation [18] delivering up to 200 mJ in 130 fs were focused p-polarized on an iron-tape target at an angle of incidence of 45°. Moreover, the target position with respect to the best-focus point was routinely optimized to have dominant K_α emission. The tape target was similar to the one described earlier [16], improved by pulling the tape over two pins close to the exit window of the target chamber (Fig. 1). In this way the capillary lens could be placed outside the chamber and was adjusted in air for optimum throughput. The capillary lens was fabricated by Institut für Gerätebau, Berlin. The front and back focal distances were 43.8 and 18 mm, respectively, and its total length was 106 mm. The radiation was detected by a charge-coupled detector (CCD), sensitized for X-rays by means of a scintillator. Visible light was blocked by a 1-mm beryllium (Be) window. The polycapillary lens has an input diameter of 3.5 mm and 2-mm output diameter protected by two 25-μm-thick Be windows at both ends. Characterization of the lens after fabrication, with an X-ray tube, shows that the lens is capable of focusing down to about 60 μm in the range 5–7.5 keV. For the alignment procedure the two Be windows were removed and the lens aligned using two counter-propagating He-Ne laser beams. The final adjustment was performed in real time running the ATLAS laser at 10 Hz and monitoring the focal spot size on the CCD using its maximum frame rate of about 1 Hz. In this way the X-ray lens can be remotely adjusted using a motorized x–y–z stage.

3 Results and applications

Correct positioning of the lens and placing the CCD in the focus of the X-ray lens and 5 mm far from it yielded the beam profiles shown in Fig. 2. These spots are recorded by irradiating the iron tape by only a single shot.

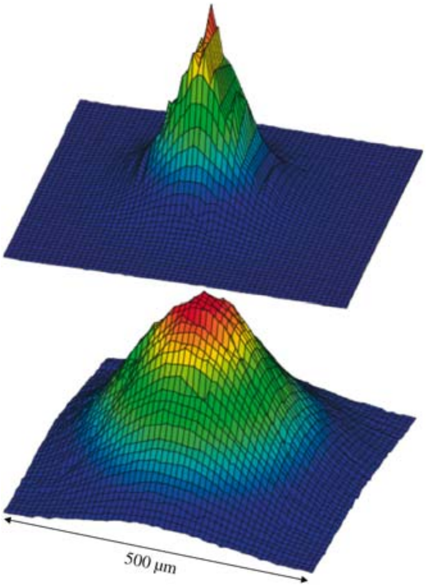

FIGURE 2 Three-dimensional beam profiles recorded by a CCD placed in the focus of the X-ray lens (*above*) and 10 mm away from the focus (*below*). The images are obtained by exposing the CCD to a single shot. In the first case the CCD counts show an enhancement of the X-ray intensity by a factor of about 1600 with respect to the case when no lens is used

The beam profiles are smoothed by the resolution of our detection system, which is comparable to the spot size at focus. In the case of the in-focus position, the enhancement of the X-ray intensity in the central peak of the focal spot was routinely found to be about a factor of 1600 with respect to the case when no lens is used. By changing the position of the CCD we measured the evolution of the cross section of the beam after the X-ray lens. The single-shot measurements of the FWHM (full width at half maximum) beam size, after deconvolution, vs CCD-to-lens distance, are shown in Fig. 3. A measured minimum beam waist of

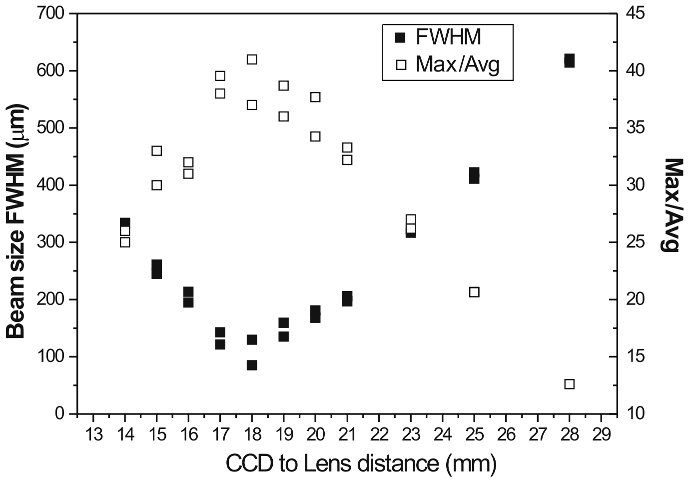

FIGURE 3 *Solid squares*: FWHM of the beam vs CCD-to-lens distance. The resulting best focus position is at 18.0 mm, with a minimum measured beam waist of 86 μm. *Hollow squares*: ratios between the maximum CCD counts and the average number of counts, i.e. a measure of the focusing of the beam

86 μm is found for a focal length of 18.0 mm. In Fig. 3 are also shown the ratios between the maximum CCD counts and the average number of counts, representing the degree of focusing of the beam. The data show a peak for a CCD-to-lens distance in very good agreement with the value corresponding to the minimum beam cross section, i.e. at 18 mm. The beam profiles after the X-ray lens are exhibited in Fig. 4 for different positions of the CCD: (a) in focus, (b) 5 mm out of focus and (c) 10 mm out of focus.

To demonstrate the application of convergent-beam ultra-short X-ray pulses for diffraction from micrometer-sized samples, a silicon crystal, oriented in the (111) direction, was placed in the focus of the X-ray lens, as shown in Fig. 1. The Bragg angle θ of the (111) reflection for Fe K_α radiation is 17.98°. A diffractogram of the focused fs-laser-generated Fe K_α radiation is shown in Fig. 5. About 300 shots corresponding to 30 s of irradiation were used to generate the pattern. In order to detect also the Fe K_β line ($\theta = 16.27°$), we deliberately overexposed the CCD with respect to the Fe K_α line (see Fig. 5). However, a well-illuminated diffractogram of the Fe K_α line with good signal to noise ratio is obtained in only 10 s.

Combining the properties of the polycapillary lens with those of the diffraction of X-rays by a crystal allows an ultra-fast streak camera to be made. The ray paths through the lens are different for different angles exiting the lens and thus the delays of different portions of the diffraction line in Fig. 5 are different. The total propagation-time difference Δt between the outermost ray and the central one is given by the geometry of the polycapillary lens and is approximated by $\Delta t = (f_1 + f_2 + l + L)(\cos(\theta)^{-1} - 1)/c$, where the symbols are defined in Fig. 6a. Substituting the values for the parameters of our setup, we obtain $\Delta t \approx 1$ ps. This is the time window of the streak camera. The temporal resolution depends on both the duration of the X-ray pulse itself and on the broadening of the pulse by propagation in a single capillary. The last contribution can be estimated by comparing the length of the capillary to the path of a ray which is totally reflected at the maximum grazing angle (termed the criti-

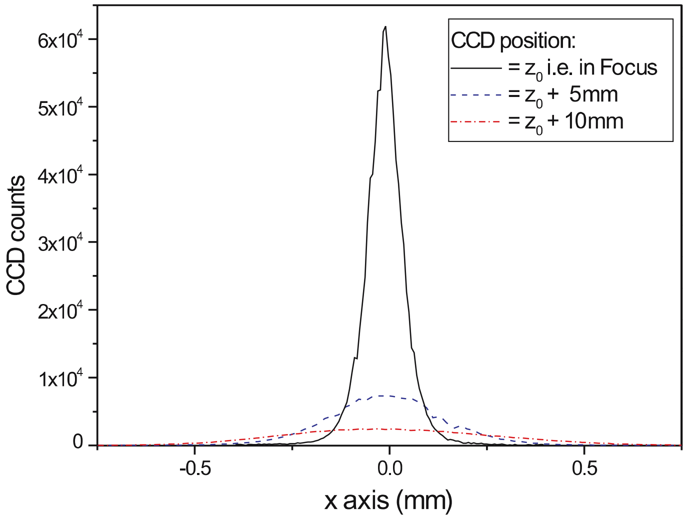

FIGURE 4 Beam cross sections at various positions: in focus and 5 mm and 10 mm out of focus, after deconvolution, for one laser shot

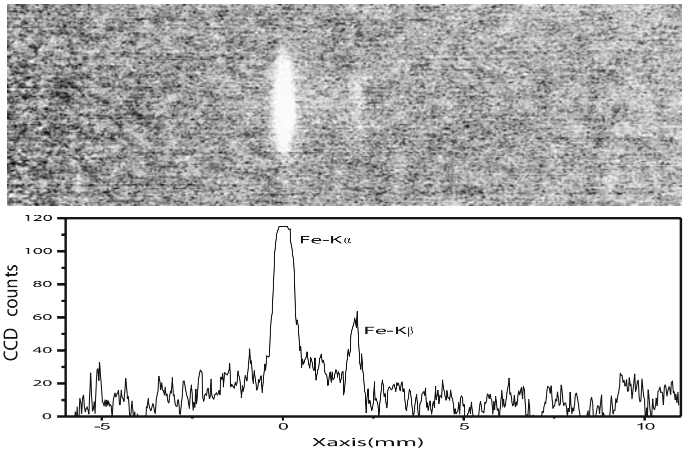

FIGURE 5 Diffractogram of the focused fs-laser-generated Fe K_α radiation: Si(111) Bragg reflex. A line-out of the diffractogram is also shown in order to emphasize the presence of the Fe K_β line, which is not so evident in the two-dimensional figure

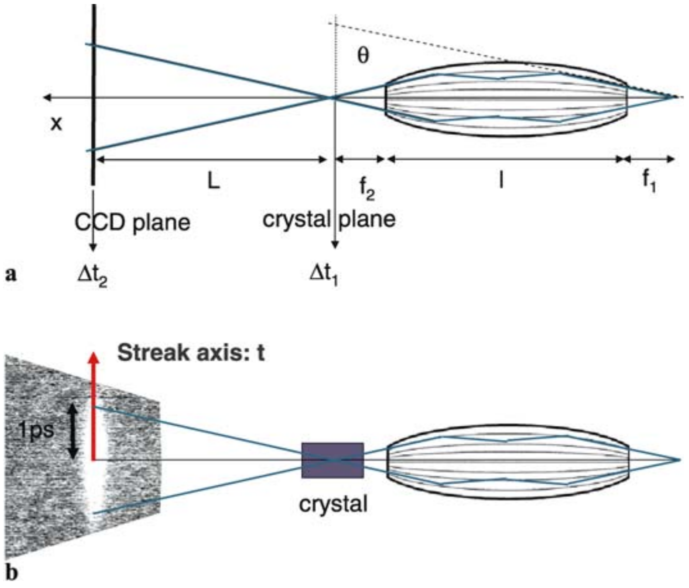

FIGURE 6 (**a**) Diagram illustrating the concept of the realization of the ultra-fast X-ray streak camera using the polycapillary lens and the diffraction from the crystal sample. (**b**) Actual setup of the ultra-fast streak camera in our experiment

cal grazing angle). The critical grazing angle of a substance is given approximately by $\Theta_{\mathrm{cr}} \approx 0.02\sqrt{\varrho}/E$, where ϱ is the density of the substance in $\mathrm{g/cm^3}$ and E is the photon energy in keV. For glass this results in a critical angle of 4.6 mrad at 6.4 keV. Thus, the intrinsic temporal broadening from propagation in one of the lens capillaries is given by $\Delta t_{\mathrm{cap}} = l(\cos(\theta)^{-1} - 1)/c \approx 3.7\,\mathrm{fs}$, which is negligibly small when compared to the pulse duration. Therefore, with our arrangement ultra-fast processes can be recorded in a 1-ps time window, as shown in Fig. 6b, with a resolution given by the duration of the pulse itself, which is about 200 fs. By modifying the lens parameters or the setup we can increase or decrease the time window. The resolution can be modified by reducing the X-ray pulse duration, i.e. by reducing the pump laser pulse duration and the tape target thickness.

4 Conclusions

In summary, we have demonstrated that a convergent X-ray beam produced by a capillary lens can be used to create a diffraction pattern with an exposure time of only a few seconds. The different delays of the various angles in the beam make it possible to use this arrangement as a novel ultra-fast X-ray streak camera and to observe the evolution of ultra-fast processes in a single run.

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