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Diffraction of laser-plasma-generated electron pulses

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ABSTRACT We report the observation of the Debye–Scherrer diffraction using electron pulses emitted from a fs-laser plasma. Titanium sapphire laser pulses with 1.6 mJ/45 fs at 1 kHz are focused on a moving steel tape at close to normal incidence. The laser plasma generated ejects a large number of electrons in the direction of polarization, with a continuous energy spectrum extending up to 100 keV. Selecting an energy range of these electrons and scattering them on a thin aluminium sample generates a "streaked" diffraction pattern with unique features.

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Time-resolved electron diffraction has recently become a powerful tool in the investigation of ultrafast processes in gases and solids [1–6]. In typical arrangements ultrashort electron pulses are generated by illuminating a photocathode with a weak laser pulse and subsequently accelerating the electrons to energies of a few tens of keV. Such experiments allow investigation of transient processes with unprecedented temporal and spatial resolution.

In this paper, we describe an alternative way of generating time-resolved electron diffractograms, i.e., using a fs-laser plasma for generating the electrons. Electrons with energies of up to about 100 keV are emitted from the plasma and therefore further acceleration is not necessary. Since the electrons generated in this way are broadly distributed in energy, the usual way of applying them would require a high degree of monochromatization. However, we show that the multi-energetic electron spectrum may constitute an advantage, since the different times-of-flight of the electrons with different energies generate an "electron streak camera" allowing investigation of fast processes in a single run.

The electron pulses are generated in the arrangement shown in Fig. 1. Pulses of a Spitfire 50 Spectra-Physics Ti:sapphire laser ($\lambda = 800$ nm, repetition rate 1 kHz, energy 1.6 mJ, pulse duration 45 fs) were focused with a lens with a focal-length of 12 cm on a slowly moving steel tape [7]. To avoid deposition of metal on the entrance window of the target chamber the angle of incidence was set at 22° from normal. The diameter of the focal spot is estimated to be 10 µm, resulting in an intensity of 4.5×10^{16} W/cm^2 on the target. The relatively low energy of the laser induces only a small indent on the tape and hence allows it to be re-used for many runs.

As a detector for the electrons an X-ray CCD (Photometrics AT200 CCD camera system with a thinned backside-illuminated chip SITe1024BP-C2) was used. The tape moved in the vertical direction and the CCD viewed the tape from the side parallel to its surface. The emitted electrons are collimated by two pinholes, the first having a diameter of 1 mm and the second, located 9 cm behind the first one, a diameter of 200 µm. The chamber was evacuated to a pressure of 2×10^{-5} mbar. The whole arrangement was encapsulated in mu-metal in order to minimize effects of the magnetic field of the earth.

The energy distribution of the electrons was determined by means of an electrostatic analyzer consisting of two 5-cm-long brass plates 1 cm apart. The deflection x of the electrons at a distance

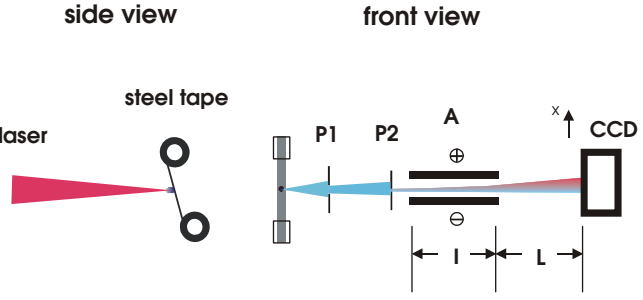

FIGURE 1 Experimental arrangement for measuring the energy distribution of electrons laterally emitted from a fs-laser plasma. **P1**, **P2:** pinholes 1 mm and 150 µm in diameter, respectively. **A:** electrostatic analyzer, plate length $l = 5$ cm, plate separation 1 cm, $L = 23$ cm. **CCD:** backside-illuminated X-ray CCD camera

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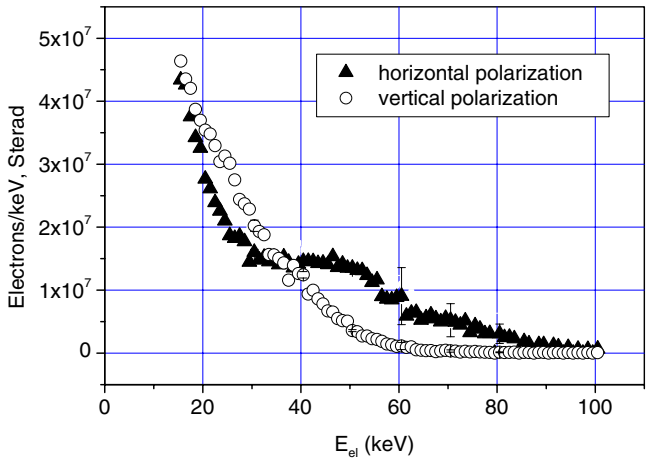

FIGURE 2 Energy distributions of the electrons emitted by a fs-laser plasma for two perpendicular polarization directions. Horizontal polarization: E-field in the direction of observation. Vertical polarization: E-field perpendicular to the direction of observation

L from the end of the plates is given by

$$x = \frac{eEl^2}{4U}\left(1 + 2\frac{L}{l}\right), \qquad (1)$$

where l is the length of the plates, e the elementary charge, E the electric field strength, and U the kinetic energy of the electrons in eV (see Fig. 1). Only about 300 ms of acquisition time was needed to obtain a well-illuminated picture. The CCD's detection efficiency has been measured for the TEK TK512M chip [8], which is equivalent to that used in our CCD camera. The results are indicated in Fig. 2 for two perpendicular polarization directions. For polarization along the direction of observation the electron energy distribution is found to extend to 100 keV with a plateau between 30 and 50 keV. For polarization perpendicular to the direction of observation, the distribution for low-energy electrons is similar, but the high-energy part is lacking.

Application of electron pulses in standard diffraction techniques requires monoenergetic electrons. Generating a quasi-monoenergetic beam by means of a slit or pinhole behind the analyzer is straightforward [9]. In this study we take a different approach, using electrons with a certain energy spread for diffraction. This "chirped" electron pulse can be applied to studies of fast changes in a sample in a single run. The principle is shown in Fig. 3: By opening the second pinhole (P2 in Fig. 3), electrons with different energies propagate along different beam paths which cross each other at a sample placed off-axis behind the analyzer. The maximum and minimum energies of electrons passing through a sample in pinhole P3 determine the temporal window of the streak camera. Their values are calculated by adding to the lateral offset x, as given by Eq. (1), the deflection due to an initial angular spread of the electron beam. The two energies are given as the solutions of the equation

$$x = \frac{eEl^2}{4U_{1,2}}\left(1 + 2\frac{L_S}{l}\right) \pm mL_0, \qquad (2)$$

where $U_{1,2}$ are the maximum and minimum electron energies, m is the initial angle from the axis, as determined by pinhole P2, L_S is the distance from the end of plates to the sample and L_0 is the distance from the source to the sample. The temporal window of the streak at P3 can then be expressed by the time difference

$$\Delta\tau = L_0(1/u_2 - 1/u_1), \qquad (3)$$

where u_1 and u_2 are the electron velocities corresponding to U_1 and U_2. The resolution of the streak camera is limited by the pixel size and is given by the time window divided by the number of pixels to which the streak extends.

A first step towards experimental realization of such an electron streak camera is demonstrated in Fig. 4a. It shows the diffraction pattern from a thin Al sample obtained with the chirped electron beam. The minimum and maximum electron energies $U_{1,2}$ were 60 and 100 keV, respectively, and the total time window was 740 ps with a resolution of the order of 20 ps. Note that the figure is rotated by 90°, such that pixel x corresponds to the vertical direction along the CCD.

A characteristic feature of the "streaked" diffractogram is its asymmetry, which is explained as follows: due to the change in electron energy along the streak, the de Broglie wavelength of the electrons, and thus the radius of the diffraction rings changes, it being larger on the right-hand side, where electrons

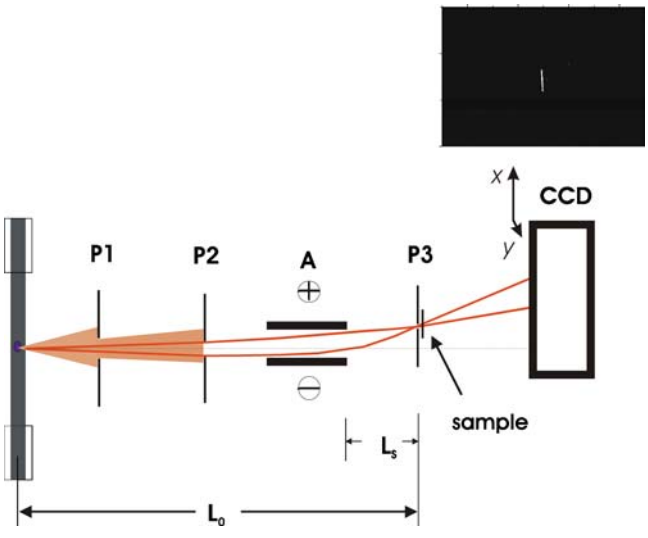

FIGURE 3 Principle and realization of a time-of-flight electron streak camera. Pinhole P2 is opened to a diameter of 1 mm. Electrons of different energies propagate along different beam paths but coincide at the sample placed immediately behind pinhole P3. $L_S = 16.5$ cm; $L_0 = 53$ cm. The CCD image on top shows the streak recorded by the CCD with no sample behind P3

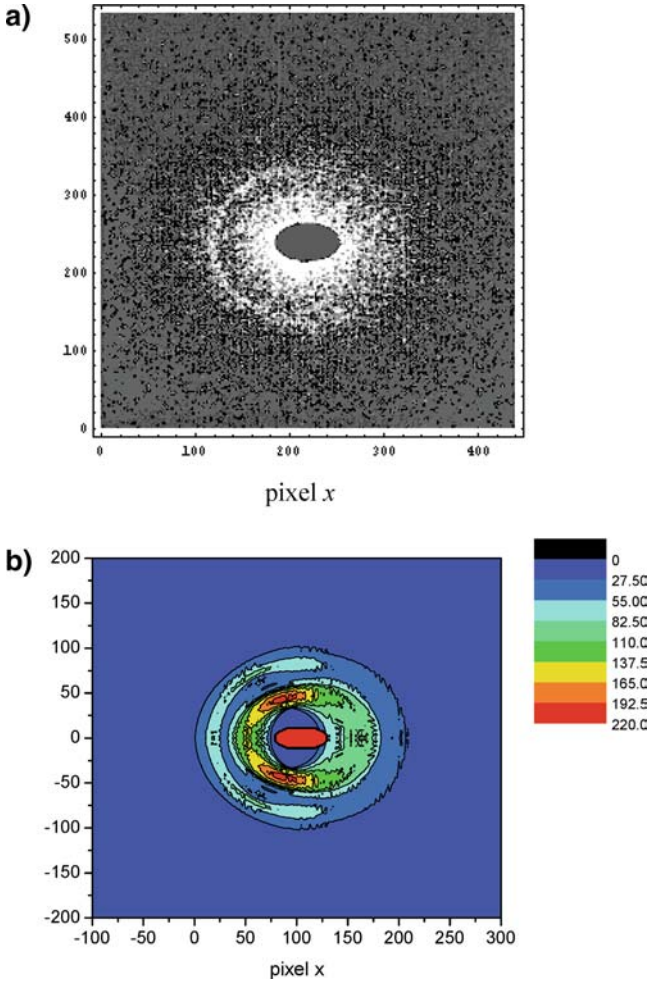

FIGURE 4 **a** Streaked diffraction pattern from a thin aluminium sample, exhibiting characteristic asymmetry. Debye–Scherrer rings coincide on the left side, owing to their different radii; **b** Simulation of a streaked diffraction pattern under the conditions of the experiment. The asymmetry of the experimental pattern is reproduced. Here "pixel x" corresponds to the vertical direction of the CCD

have a lower energy. Therefore on the left side the rings partially coincide, resulting in a sharp ring structure, while on the right side of the pattern they are smeared over a larger area. The simulation of such a "streaked" Al diffraction pattern is shown in Fig. 4b. This Al diffraction spectrum was obtained by means of the PowderCell diffraction code [10], modified to take dynamic diffraction effects into account. An elliptic electron beam with an energy range between 60 and 100 keV was assumed to be scattered on a polycrystalline aluminium sample. The four strongest allowed diffraction lines associated with a face-centered cubic lattice, namely (111), (200), (220) and (311) are taken into account. The resulting pattern clearly reproduces the asymmetry of the experiment.

The method of "streaked electron diffraction" seems well suited for studies of fast processes in matter. As an example, we mention here conformational changes in organic molecules, which generally occur on a ps-time scale [11, 12]. Compared to X-rays the large scattering cross-section of electrons allows much thinner samples to be used for generating a diffraction pattern, resulting in an ideal matching of the excitation and probe depths. Moreover, the larger scattering cross-section also results in shorter acquisition times and therefore lower dose for recording a specific pattern. Another advantage is that due to their weaker Z-dependence of the scattering cross-section electrons respond more effectively to changes in lighter systems with few or no electrons, such as H-atom and proton positions, a feature most welcome in investigations of organic materials. In this context we note that a "chirped" electron pulse is not affected by broadening due to electron-electron Coulombic repulsion and therefore, the achievable temporal resolution is not limited by this effect.

In summary, we have shown that electrons emitted from a fs laser plasma can be used in diffraction experiments. Such investigations using electrons in a certain energy range may explore transient processes in a single series of laser shots, taking advantage of the different arrival times of the electrons on a sample. A surprising feature of the chirped electron pulse technique is that it can act as a "time magnifier" upon propagation: For example, a 60 keV electron pulse with an energy spread of 10%, broadens by about 3 ps for each centimeter of propagation. Thus, rapid changes early in the beam path are mapped onto a much larger time window after a certain distance, displaying the temporal evolution in slow motion.

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