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Generation of monoenergetic ultrashort electron pulses from a fs laser plasma

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ABSTRACT Ultrashort high-energy electron beams are generated by focusing fs Ti:sapphire laser pulses on a thin metal tape at normal incidence. At laser intensities above 10^{16} W/cm^2 , the fs laser plasma ejects copious amounts of electrons in a direction parallel to the target surface. These electrons are directly detected by means of a backside illuminated X-ray CCD, and their energy spectrum is determined with an electrostatic analyzer. The electrons were observed for two laser polarization directions, parallel and perpendicular to the observation direction. At the maximum applied intensity of 2×10^{17} W/cm^2, the energy distribution peaks at around 35 keV with a hot tail detectable up to about 300 keV. The number of electrons per shot at 35 keV is about 5×10^8 per sterad per keV. Quasi-monoenergetic electron pulses with a relative energy spread of 1% were produced by using a 50-μm slit in the beam path after the analyzer. This approach offers great potential for time-resolved studies of plasma, liquid, and surface structures with atomic-scale spatial resolution.

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The availability of Terawatt laser systems with a pulse duration in the fs range and of 10-Hz repetition rate has opened up the possibility of powerful laser-target interaction experiments to investigate numerous aspects in the generation of X-rays, energetic electrons and ions [1–7]. X-ray and electron diffraction combined with ultrafast temporal resolution can provide unique capabilities for advanced structural investigations. While laser-plasma generated X-ray pulses are widely used for investigating transient phenomena, laser-plasma generated electrons do not seem to have been applied for this purpose. Comparison of the two methods shows electron diffraction to have the advantage that the scattering cross-section is many orders of magnitude larger than those for X-rays. Electron diffractograms can, therefore, be obtained with a much lower dose of incident radiation, and in a pump-probe experiment the depth of excitation and the probe depth can be ideally matched to each other. Furthermore, this method is more surface-sensitive and allows ultrashort measurements of crystals, surfaces, and macromolecules with unprecedented spatial resolution.

Electron diffraction by ps electron pulses has been demonstrated by Zewail and coworkers [8–10]. In their work, the electrons were generated by laser-induced emission from a photocathode and subsequent acceleration by an applied high voltage. Ultrafast electron diffraction on gaseous and solid samples has been demonstrated by this method. For example, the temporal evolution of the stepwise elimination of two iodine atoms from $C_2F_4I_2$ by the photodissociation reaction, $C_2F_4I_2 \rightarrow C_2F_4 + I + I$, was observed in the gaseous phase. This would have been impossible by other methods, such as X-ray diffraction or infrared absorption.

In this work, we report the observation of intense ultrafast electron emission from a fs laser plasma without any further acceleration, thus having the advantage of compactness and simplicity. We demonstrate for the first time that monoenergetic ps or fs electron pulses can be laser-generated with a relatively straightforward arrangement. We note that electrons generated by intense laser pulses are broadly distributed in energy and thus could in principle be used for diffraction experiments using single crystals in a Laue configuration. However, to our knowledge this possibility has hitherto never been used in experiments and is under investigation by our group. Here it is shown that the number of laser-generated hot electrons in the 20–50 keV region is high enough for monochromatization and use in future diffraction experiments in polycrystalline materials.

The experimental set-up for ultrafast electron generation is shown in Fig. 1. Pulses of a Ti:sapphire laser ($\lambda = 86$ nm, repetition rate 10 Hz, energy 150 mJ, pulse duration 150 fs) were focused by a lens of 60 cm focal length on a slowly moving copper tape at normal incidence. The diameter of the focal spot is about 25 μm, resulting in a maximum intensity of 2×10^{17} W/cm^2 on the target. The measured prepulse ASE level of the laser was 10^{-7} for 2 ns ahead of the main pulse and about 10^{-8} for roughly 5 ns before the main pulse [11].

The continuous tape target is similar to that previously described in the context of our X-ray generation experiments [12]. A 10-m-long and 12-mm wide copper tape is pulled across the

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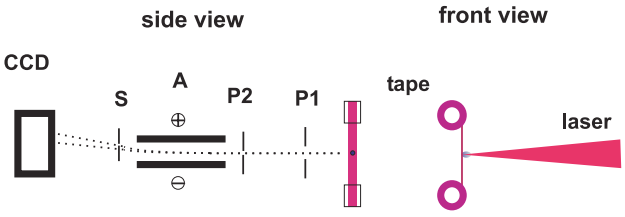

FIGURE 1 Experimental arrangement for detecting and analyzing electrons emitted from a fs laser plasma. P1, P2: pinholes 1 mm and 200 μm in diameter, respectively. A: electrostatic analyzer, plate length 5 cm, plate separation 1 cm. S: 50-μm slit for generating quasi-monoenergetic electrons. CCD: backside-illuminated X-ray CCD-camera. *Dotted lines* show paths of electrons for two different energies

laser focus at a velocity of about 1 cm/s, making it possible to accumulate up to 10 000 shots in a single run. By slightly displacing the lens, a single tape can be used for many runs, making this target technique easy and cost-effective to operate.

An X-ray CCD (Photometrics AT-200 CCD camera system with a thinned backside-illuminated chip SITe-1024BP-C2) was positioned in the plane of the target surface, about 60 cm from the target. The tape moves in a vertical direction and the CCD viewed the thin side of the tape, i.e., the observation is in the plane of the target surface. In this geometry the laser pulse is reflected out of the target chamber and illumination of the CCD by laser light is minimized. A low-divergence beam is generated by passing the emitted electrons through 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 energy distribution of the electrons is determined by means of a simple electrostatic analyzer (for details see Fig. 1). The deflection angle of an electron in a homogeneous electrostatic field is given by $\tan^{-1}(lE/2U)$, where l is the length of the plates, E the field strength and U the kinetic energy of the particle in eV when entering the analyzer. Figure 2 shows a typical example of a two-dimensional CCD readout of the electron spots corresponding to 30 laser shots. The spots are arranged along a parabola, due to the electrostatic field (deflection in the x-direction) and the magnetic field of the earth (deflection in the y-direction).

Electron energy distributions are determined for different laser parameters, such as intensity and polarization. The response of the CCD to electrons of different energies was calibrated by correlating the number of counts to the distance of deflection of the electrons by means of the electrostatic field analyzer. To prevent distortion of the data due to event-splitting or charge migration [13] only single-pixel events were used for this purpose. Multiple illumination of pixels was minimized by using for this evaluation a CCD image as in Fig. 2, but generated by only a single shot. A linear relation was found between the electron energy and the number of counts per electron on the CCD. The number of incident electrons is then calculated using the CCD's detection efficiency function as given for the TEK TK512M chip in [14]. This chip is equivalent to that used in our CCD. The resulting distributions were normalized with respect to an energy interval of 1 keV and a solid angle of 1 sterad.

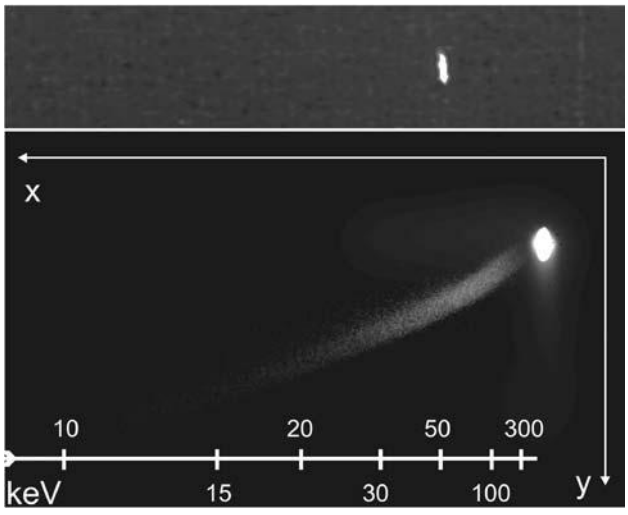

FIGURE 2 CCD image of the electrons emitted by the fs laser plasma (30 shots). The *image at the top* shows a monoenergetic electron beam transmitted through the slit S (see Fig. 1). To obtain the *image below*, the slit was removed. The dispersion in the x-direction is due to the electric field (here 200 V/cm) in the analyzer, while the dispersion in the y-direction is due to the magnetic field of the earth (the picture is rotated by 90°). The energy scale is displayed at the bottom. The vertical width of the structure corresponds to the beam diameter (defined by the 200-μm pinhole, see Fig. 1). The *bright spot* at the upper right of the lower picture is produced by X-rays. It is eliminated if the slit S is in place

Figure 3 shows our results for different intensities and polarizations of the fs laser pulse and for a single laser shot. The data below 20 keV are not displayed because the pattern of low-energy electrons on the CCD is broadened by stray fields, resulting in a large error bar. For shots with maximum intensity (2×10^{17} W/cm^2), the energy distribution peaks at about 35 keV and the detectable distribution extends up to 300 keV. For lower irradiance, the total number of electrons is significantly reduced but the peak only slowly shifts to lower energies. However, we note that even for an energy as small as 20 mJ (corresponding to an intensity of 2.7×10^{16} W/cm^2) we still measure about 6×10^7 electrons/(keV sterad) per shot, at an energy of 20 keV. Measurements corresponding to 10 mJ of laser energy (not shown in the figure for the sake of clarity) show a number of generated electrons only a factor of two lower. This demonstrates the feasibility of our method using much weaker laser drivers and thus the possibility to increase the repetition rate.

For application of the generated electron beam in time-resolved diffraction experiments, the electrons need to be monochromatized. To this end, a slit is used in an arrangement as shown in Fig. 1. Given the relation, $U \propto x^{-1}$, between the energy of the electrons, U, and the lateral displacement, x, due to the electrostatic analyzer, the rela-

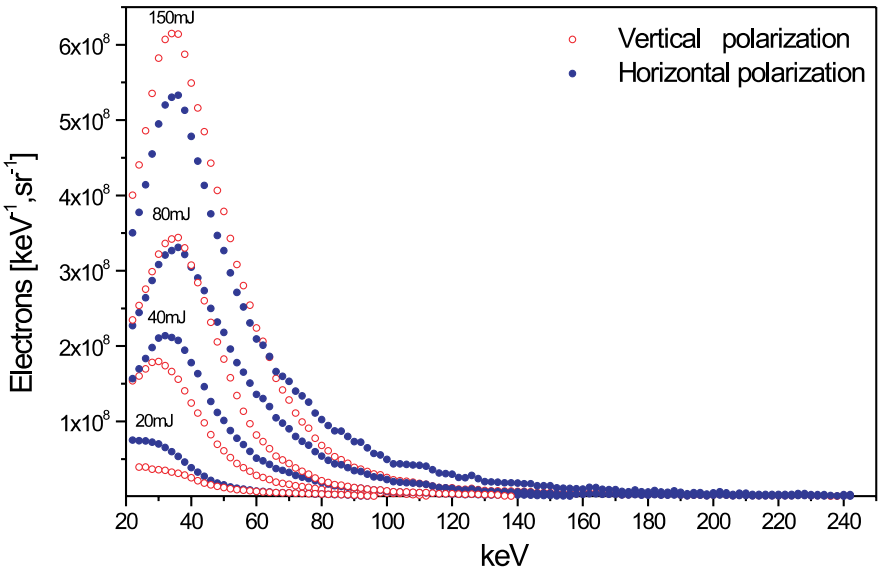

FIGURE 3 Energy distributions of the electrons emitted by a fs laser plasma, for different laser energies and horizontal and vertical polarization directions, i.e., observation parallel and perpendicular to the electric vector of the laser light

tive energy width of the electron beam after passing the slit is simply given by $\Delta U/U = \Delta x/x$, where Δx is the slit width. The energy spread thus depends only on the geometry. In our case, a 50-μm wide slit was inserted between the analyzer and the detector, 5 mm off axis, resulting in a relative energy width of 1% for the electron beam. A CCD image of the electrons emitted by the fs laser plasma (30 shots) and monochromatized by the slit is shown in the upper part of Fig. 2.

We note that due to the energy width the beam exiting the slit has a defined pulse duration. Consider a sample positioned behind the slit, at a distance d from the electron source. The arrival time of electrons having speed, v, is given by $t = d/v$. Due to the energy width, the electrons arrive on the sample during the time interval, $\Delta t = t\Delta v/v = (t/2)\Delta U/U$. Hence the pulse duration of the electron beam impinging on the sample is given by $\tau_s = (d/2v)\Delta x/x$. In our experimental setup, the pulse duration for electrons with 35 keV, immediately behind the 50-μm slit, is about 22 ps. Note that this pulse duration can easily be reduced to the the ps or sub-ps range by modifying the relevant geometric parameters, i.e. the slit width, its off-axis displacement and the sample-source distance. In the sub-ps range, the above relation must be corrected to take into account the time of emission of the electron source itself, which depends on the pulse duration, τ_L , of the fs laser. The true pulse duration of the electron beam can be approximated by $\tau = \left(\tau_s^2 + \tau_L^2\right)^{1/2}$.

In designing an electron diffraction experiment with a particular time resolution, one would first select the appropriate energy width. Then, from the data of Fig. 3 one can calculate the number of electrons in the beam. In the particular experiment reported here, considering the data for 150 mJ of pump laser energy and vertical polarization (see Fig. 3), the 50-μm wide slit selects a beam with an energy width of 1%, centred at 35 keV, a pulse duration of 22 ps and a total number of electrons $\approx 2.13 \times 10^8$ persteradpershot. Experiments for investigating fast conformational changes in the organic crystal DIABN (diisopropylamino-benzonitrile, $C_{13}H_{18}N_2$) using such electron pulses are in preparation.

We note that the physics underlying the lateral emission of electrons from a laser plasma with energies well above the ponderomotive potential is not fully understood. Chaotic electron acceleration in the field of a standing wave is suggested as a possible mechanism [15, 16]. The difference in the distributions for the two polarization directions is puzzling. Model calculations including the ponderomotive force and taking the transverse and longitudinal fields of the focused laser pulse into account are in progress.

In summary, we have demonstrated that a sufficient number of electrons are emitted in the target plane of a fs laser plasma to monochromatize them for planned diffraction experiments and other studies. Our approach to effectively producing fast electrons in the 20–60 keV range can be exploited, e.g., for laser plasma studies as well as to probe transient reactions on a femtosecond or picosecond time scale, phase changes in solids or conformational changes in proteins. In addition, such short electron pulses could be used for time-resolved electron microscopy.

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