Femtosecond electron pulse characterization using laser ponderomotive scattering

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We demonstrate a method for the measurement of the instantaneous duration of femtosecond electron pulses using the ponderomotive force of an intense ultrashort laser pulse. An analysis procedure for the extraction of the electron pulse duration from the transient change of the transverse electron beam profile is proposed. The durations of the electron pulses generated in our setup were determined to be 410 ± 30 fs.

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Ultrashort electron pulses have generated interest in recent years for their applications in femtosecond electron diffraction (FED) (Ref. 1 and references therein) and time-resolved electron microscopy.[2] The hybrid approach of an optical pump and electrons as a probe has enabled the direct observation of atomic motion in real time and is providing first direct glimpses at transition state processes. These processes take place on the 100 fs time scale. The pulse used to probe these events must be comparably short or shorter in duration. Many structural changes that can be examined with FED are nonreversible, and hence it is also imperative to maximize the available diffraction signal per pulse by using a large number of electrons per pulse. It remains a great experimental challenge to generate such electron pulses on this time scale. A method that is capable of reliably characterizing the duration of electron pulses is therefore essential.

Ultrashort electron pulses are usually generated by photoactivated electron guns. In these electron pulse sources, a photocathode is illuminated by a femtosecond laser pulse,[3] and the resulting electron cloud is accelerated by a DC electric field over a potential difference of tens of kilovolts. The initially generated electron pulse mirrors the duration of the laser pulse. The excess photon energy above the work function of the photocathode leads to an initial kinetic energy distribution of the electrons. More important, electrostatic forces between the electrons make them repel each other. This space-charge effect leads to a rapidly changing pulse duration as the electron pulse propagates.

The conventional method for measuring the duration of electron pulses is to apply a ramped electric field normal to the propagation direction. This streak camera technique is sufficient for picosecond pulses and can—for small low-density electron beams—be extended into the femtosecond regime.[4] However, there are problems with streak cameras that limit their ability to measure high-number-density femtosecond electron pulses: a short pulse also creates a short streak on the detector. When the length of the streak is of the same order of magnitude as the unstreaked spot size, the pulse duration cannot be recovered reliably. More important, the aforementioned space-charge broadening effect can lead to significant increases of the pulse duration over propagation distances of the order of a few centimeters (the typical length of streaking plates) for typical electron densities, rendering the pulse duration ill defined.

To obtain a reliable measurement of the instantaneous pulse duration, we proposed using the laser ponderomotive force to sequentially scatter parts of the electron beam.[5] In this technique, the electron beam path is overlapped with the focus of an intense femtosecond laser pulse. While the force of a continuous plane wave on a charged particle averages out over a cycle, this is not the case for the highly inhomogeneous field of a focused laser pulse. The ponderomotive force that the electrons experience in this field is approximately described by[6]

$$\mathbf{F}(\mathbf{r},t) = -\frac{e^2\lambda^2}{8\pi^2 m_e \epsilon_0 c^3} \nabla I(\mathbf{r},t). \tag{1}$$

Here, e and m_e are the charge and mass of an electron, respectively, λ is the wavelength of the laser, and I is the laser intensity. To our knowledge, other proposals to use the ponderomotive force for measuring the duration of electron pulses by using evanescent waves[7] or surface plasmons[8] have not been demonstrated experimentally. A previous experimental investigation[9] used extreme laser intensities requiring a relativistic treatment of the ponderomotive interaction.

The experiments presented here were performed using the beam line 2B of the Advanced Laser Light Source, Varennes, QC, Canada. This beam line operates at 100 Hz and provides pulses up to 40 mJ with a bandwidth of 40 nm centered around 800 nm. The results were obtained by using laser pulses stretched to 90 fs with a pulse energy of 14.9 mJ focused to a spot size of 11.9 ± 0.3 μm FWHM leading to a peak intensity of 9.7×10^{16} W/cm^2. The electron gun was operated at $V = 55$ kV at a fluence of 4×10^7 electrons/cm^2 and with a 100 μm diameter pinhole.

The experimental setup is shown schematically in Fig. 1. Tripled light (267 nm) used to create the electron pulse is produced by two β-barium borate crystals cut for type I second-harmonic generation (SHG) and type II sum-frequency generation (SFG), respectively. A prism compressor compresses the resulting UV pulse and separates the tripled light from the fundamental and doubled light. Spatial overlap of the laser and electron beams is achieved by using an elliptical aperture tilted by 45° relative to both beams.

A series of images of the electron spot on the detector at different delays is acquired by using a cooled CCD camera (see Fig. 2). The time resolution of this experiment is limited by the duration of the laser pulse and the mutual crossing times of the laser and electron pulses. To improve the resolution, we strip the electron beam of its outer electrons by using a pinhole immediately before the beam crossing. This leads to a trade-off between temporal resolution and the number of detected electrons.

While the number of frames that contain significant distortion of the electron beam can serve as an estimate for the duration of the electron pulse, for an accurate measurement it is essential to find a quantity that is related to the profiles of the electron pulse and the laser pulse in a simple way, as shown below.

The laser pulse propagates along the x-axis in the positive direction, and the electron pulse propagates along the z-axis also in the positive direction at velocity v. Because the size of the laser focus is small compared with its distance from the detector, the change of the position of an electron in the detector plane due to deflection by the laser ponderomotive force can be written as

$$[X,Y] = \frac{T}{m_e}\int [F_x(\mathbf{r}(t),t), F_y(\mathbf{r}(t),t)]\mathrm{d}t. \qquad (2)$$

Here, T is the time the electrons take to propagate from the scattering region to the detector, and $\mathbf{F}$ is the force the electron experiences at its position $\mathbf{r}(t)$

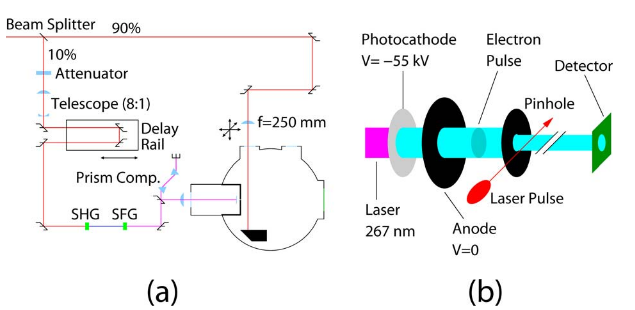

Fig. 1. (Color online) (a) Optical setup of the experiment. The incoming laser pulse is split into two arms: 90% of the pulse energy is focused to scatter the electron beam, and the remaining 10% is further attenuated, sent through a variable delay line, tripled, and compressed before entering the electron gun. (b) Schematic view of the experiment geometry. When the tripled laser pulse hits the photocathode, it creates an electron pulse that is accelerated by a DC electric field. The electron pulse is then stripped of its outer electrons by a pinhole before it is scattered by the ponderomotive force of the laser pulse. The electrons are detected by a microchannel plate/phosphor detector.

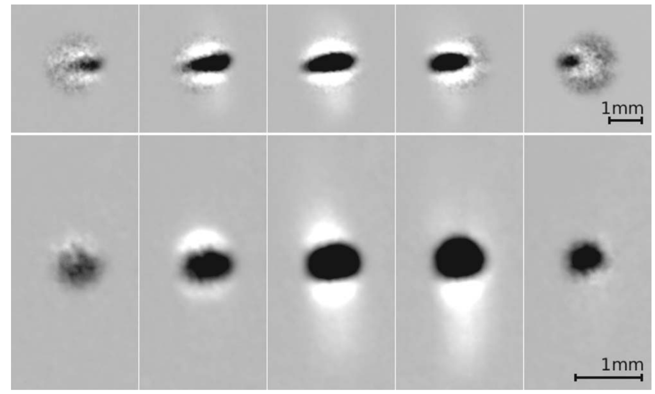

Fig. 2. Series of images of the beam on the detector without (upper row) and with (lower row) the stripping pinhole (30 μm) in place. An image of the undisturbed beam has been subtracted from each of the images to highlight the scattering of electrons due to the ponderomotive force of the laser pulse. The time step between adjacent images is 300 fs.

due to the laser field. The force in the z-direction has a negligible effect because of the high initial velocity of the electrons in this direction. For a short time around the overlap of the electron and laser pulses, let us assume that the envelopes of both pulses are constant. They are described by the number density of the electron pulse $n(x,y,z-vt)$ and the ponderomotive force profile of the laser pulse $\mathbf{F}(x-ct,y,z)$. According to Eq. (1), the latter is proportional to the intensity gradient, and hence the force in the y-direction is proportional to the temporal laser pulse envelope. The deflection of an electron incident at $(x,y,z-vt)$ in the y-direction in the detector plane is

$$Y = \frac{T}{m_e}\int F_y(x-ct,y,z+vt)\mathrm{d}t \qquad (3)$$

as long as the lateral motion of the electron while crossing the laser focus is negligible.

As an experimentally accessible signal, we choose the product of the absolute value of this vertical displacement and the detected electron number density $D(X,Y)$ integrated over the detector area as a measure of the ponderomotive scattering:

$$S_{\mathrm{ponderomotive}} = \int |Y| D(X,Y)\mathrm{d}X\mathrm{d}Y = \frac{T}{m_e}\int n(x,y,z) \times |F_y(x-ct,y,z+vt)|\mathrm{d}t\mathrm{d}x\mathrm{d}y\mathrm{d}z. \qquad (4)$$

The force profile of the laser beam can be rewritten as $|F_y| = F_0 f_t(x/c) f_y(y) f_z(z)$ for a Gaussian pulse assuming Eq. (1). If the laser focus is small compared with the electron beam diameter, the spatial profile of the electron beam can be approximated as $n(x,y,z) = N_0 n_x(x) n_y(y) n_t(z/v)$ in the interaction volume. If we introduce a delay τ in the laser pulse, we get

$$S_{\text{ponderomotive}}(\tau) = \frac{N_0 F_0 T}{m_e} \int f_y(y) n_y(y) \mathrm{d}y \times \int \int n_x(x) f_t\left(\frac{x}{c} - t + \tau\right) \mathrm{d}x \times \int n_t\left(\frac{z}{v}\right) f_z(z + vt) \mathrm{d}z \mathrm{d}t, \qquad (5)$$

which is easily recognized as a convolution of $f_z(vt)$, $n_x(-ct)$, $f_t(t)$ and the sought-after temporal profile of the electron pulse $n_t(t)$. Hence the signal can be viewed as a cross correlation between the temporal pulse shapes of the laser and electron pulses with additional broadening caused by the spatial pulse shapes.

We reduce the noise caused by shot-to-shot fluctuations of the electron beam current by normalizing the images with respect to their total intensity. Signal traces and their respective Gaussian fits are shown in Fig. 3. The results were obtained with approximately 3100 electrons/pulse in a beam diameter of 100 μm.

To recover the pulse duration from these traces, the other contributions have to be determined. As the stripping pinhole is much smaller than the diameter of the electron beam, the resulting stripped electron beam should not have any notable structure normal to the propagation direction and can be treated as a top-hat profile with its width equal to the size of the pinhole (30 μm). The spatial and temporal envelopes of the laser pulse are assumed to be Gaussian. The convolution of these contributions is approximated well by a Gaussian with a FWHM of 147 fs. Hence, from the measurement of the stripped beam, we can determine the pulse duration of the electron pulse to be 410 ± 30 fs.

Since the determined pulse duration is very close to the width of the measured trace, this measurement should be largely independent of the exact shape of the spatial contributions to the trace. In contrast, the measurement with the complete electron beam can at best deliver a rough estimate of the pulse duration unless the spatial shape of the electron pulse, which dominates the duration of the signal, is known with good accuracy. By further reducing the various contributions to the system response and increasing the signal-to-noise ratio, it should be possible to not only determine the pulse duration to better than 100 fs resolution but also to measure the temporal profile of the electron pulse with this method. With an improved signal-to-noise ratio, a pulse energy of about 1 mJ as obtained from a conventional Ti:sapphire laser system should be sufficient to perform this electron pulse characterization. We also note that this method is capable of determining the temporal overlap ($t=0$ position) for FED experiments to better than 100 fs accuracy.

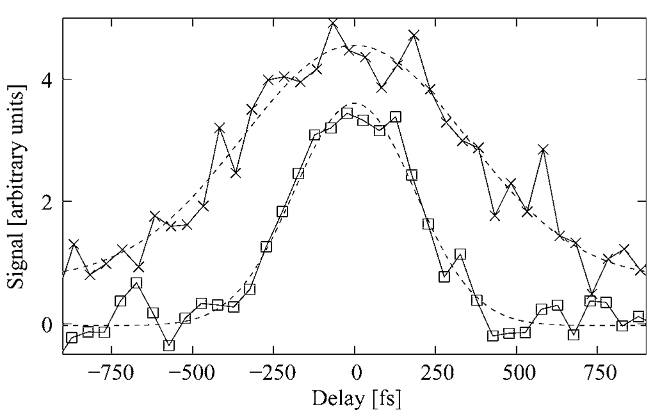

Fig. 3. Signal traces without (crosses) and with (squares) stripping pinhole. The FWHM of the Gaussian fits (dashed curves) are 815 ± 49 fs and 438 ± 22 fs, respectively.

In conclusion, we demonstrated a method to measure the durations of ultrashort electron pulses on the order of hundreds of femtoseconds. This method solves the previously outstanding issues with respect to electron pulse duration and temporal overlap. This is essential for FED experiments to achieve the highest time resolution possible to atomically resolved structural dynamics.[1,10] We also showed that pulses produced by our setup with an electron fluence of 4×10^7 electrons/cm^2 and energy of 55 keV are just over 400 fs long.

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