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DOI:
10.1103/PhysRevLett.105.264801

Document status and date:
Published: 01/01/2010

Document Version:
Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Compression of Subrelativistic Space-Charge-Dominated Electron Bunches for Single-Shot Femtosecond Electron Diffraction

T. van Oudheusden, P. L. E. M. Pasmans, S. B. van der Geer, M. J. de Loos, M. J. van der Wiel, and O. J. Luiten*

Department of Applied Physics, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands

(Received 6 June 2010; published 22 December 2010)

We demonstrate the compression of 95 keV, space-charge-dominated electron bunches to sub-100 fs durations. These bunches have sufficient charge (200 fC) and are of sufficient quality to capture a diffraction pattern with a single shot, which we demonstrate by a diffraction experiment on a polycrystalline gold foil. Compression is realized by means of velocity bunching by inverting the positive space-charge-induced velocity chirp. This inversion is induced by the oscillatory longitudinal electric field of a 3 GHz radio-frequency cavity. The arrival time jitter is measured to be 80 fs.

DOI: 10.1103/PhysRevLett.105.264801 PACS numbers: 41.75.Fr, 29.27.−a, 52.59.Sa, 61.05.J−

The breathtaking pace at which ultrafast x-ray and electron science have evolved over the past decade is presently culminating in studies of structural dynamics with both atomic spatial *and* temporal resolution, i.e., sub-nm and sub-100 fs [1–3]. This may revolutionize (bio) chemistry and materials science and may even open up completely new areas of research. A particularly exciting development is the hard x-ray free electron laser, which became operational in 2009 [3]. This has already resulted in single-shot, femtosecond x-ray diffraction experiments on submicron crystals of a membrane protein [4]. In parallel ultrafast electron diffraction (UED) techniques have been successfully applied to investigate the structural dynamics of, in particular, condensed matter phase transitions [5–8]. X-ray diffraction and electron diffraction provide principally different, but in fact complementary, information of atomic structure. Because of their shorter mean free path, however, electrons are favorable for probing thin films, surfaces, and gases. Unfortunately, single-shot, femtosecond operation has not yet been achieved with electrons: because of the repulsive Coulomb force high charge density bunches will expand rapidly in all directions. By accelerating the electrons to relativistic speeds as quickly as possible the Coulomb force is damped, thus slowing down the expansion. Although relativistic bunches can be used for UED [9] they pose difficulties like a reduced cross-section, radiation damage, and nonstandard detectors. Electron energies of 100–300 keV are generally preferred [10], but inevitably result in loss of temporal resolution. The obvious solution is to lower the charge per bunch [2,5,6] and use multiple shots to obtain a diffraction pattern of sufficient quality. However, in this way the choice of samples is restricted for reasons of radiation damage and repeatability of the process under investigation. The closest approach to single-shot, femtosecond operation has been achieved by Sciaini *et al.*, who used $\sim$0.001 pC bunches and integrated 4–12 shots per time point to monitor electronically driven atomic motions of Bi [6]. By positioning the sample at 3 cm from the photocathode they achieved a full-width-at-half-maximum (FWHM) bunch duration $\gtrsim 350$ fs. Preferably, however, the bunch charge should be $\gtrsim 0.1$ pC, in particular, for UED on more complicated molecular crystals, while maintaining a high beam quality and $\lesssim 100$ fs resolution.

In this Letter we demonstrate 100-fold compression of 0.25 pC electron bunches to sub-100 fs root-mean-square (rms) durations (Fig. 1). These bunches are of sufficient quality to record the diffraction pattern of a polycrystalline gold foil in a single shot (Fig. 2).

The quality of a diffraction pattern is mainly determined by the transverse coherence length L_c of the electron bunch, defined as $L_c = \lambda/(2\pi\sigma_\theta)$, where λ is the electron de Broglie wavelength, and σ_θ the transverse rms angular spread of the electrons. The transverse coherence length should preferably be larger than the lattice spacing, implying $L_c \gtrsim 1$ nm. Further, the transverse beam size should be matched to the size of the crystal sample, which is often

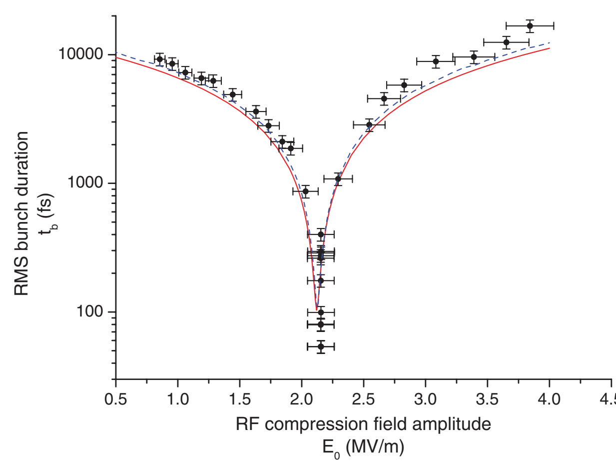

FIG. 1 (color). rms duration t_b of 95.0 keV, 250 fC electron bunches measured as a function of the rf compression field amplitude $E_0(0)$ (black dots). GPT simulations, both with (blue dashed line) and without (red solid line) slit, are in close agreement with the measurements.

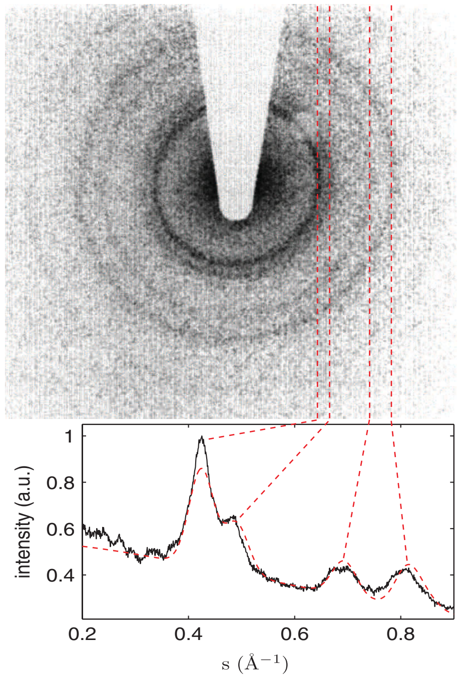

FIG. 2 (color). (top) Electron diffraction pattern of a polycrystalline gold foil, recorded with a single 200 fC bunch. (bottom) Azimuthal integration of Debye-Scherrer rings (black line) and a fit (red dashed line) based on kinematical diffraction theory.

limited to $\lesssim 100\ \mu$m. From the requirements on L_c and on the beam size at the sample it follows that the femtosecond photoemission laser spot size on the photocathode should be $\lesssim 100\ \mu$m [10]. For a high-quality diffraction pattern at least 0.1 pC ($\sim 10^6$ electrons) is required. Under these conditions the initial bunch dynamics are dominated by strong and generally nonlinear space-charge forces. As a result the bunch will not only expand rapidly, but it will also deteriorate: the rms angular spread (measured in the beam waist) will increase, leading to a smaller L_c. By carefully shaping the transverse intensity profile of the photoemission laser pulse an ellipsoidal bunch with a homogeneous charge distribution can be realized [11]. The expansion of such a bunch is fully reversible with linear charged particle optics; i.e., L_c is not affected by space-charge forces. Reversal of the transverse expansion can then be realized with regular charged particle lenses; for reversal of the longitudinal expansion there is, however, no standard method available. In Ref. [10] we proposed the use of radio-frequency (rf) techniques. We investigated the feasibility of the rf bunch compression method by detailed particle tracking simulations, including space-charge effects and realistic external fields of the charge particle optics. Here we present the experimental realization of this method, with the emphasis on achievable bunch length and the application to UED. For a more detailed discussion of the underlying beam physics we refer to Ref. [10].

Our setup is schematically shown in Fig. 3 and is described in more detail in Ref. [10]. A ≥ 0.1 pC bunch is created by photoemission with the third harmonic of a 50 fs, 800 nm laser pulse. The transverse laser profile is Gaussian with a 200 μm waist, concentrically truncated by a 200 μm diameter pinhole. The resulting dome-shaped surface charge distribution has been demonstrated to evolve during acceleration into an ellipsoidal bunch [12,13].

After photoemission the rapidly expanding electron bunch is accelerated in a 112 kV/cm dc electric field to an energy of 95 keV. Two solenoids control the transverse bunch size. To compress the bunch in the longitudinal direction we use a rf cavity oscillating at $f = 3$ GHz in the TM_{010} mode. The oscillating on-axis longitudinal electrical field $E_z(z, t) = E_0(z)\cos(2\pi f t)$ decelerates the electrons at the front of the bunch and accelerates the electrons at the back of the bunch, leading to velocity bunching in the subsequent drift space. Maximum compression is achieved for an electric field strength at the center $E_0(0) =$ 2.2 MV/m (Fig. 1), which requires only 51 W of rf power owing to the optimized shape of our cavity [10].

To measure the bunch length a 3 GHz rf cavity oscillating in the TM_{110} mode is employed, which acts as an ultrafast streak camera [14]: the oscillating on-axis perpendicular magnetic field $B_y(z, t) = B_0(z)\cos(2\pi f t)$ deflects the electrons in the x direction. The rf phase is chosen such that the electrons at the center of the bunch are not deflected. In this way the longitudinal bunch profile is projected as a streak onto the xy plane. For detection we use a microchannel plate (MCP) with a phosphor screen that is imaged 1:1 onto a CCD camera. The relation between the bunch duration t_b and the length X_{str} of the streak on the phosphor screen is obtained by integrating the Lorentz force on the bunch. For $t_b \ll f^{-1}$

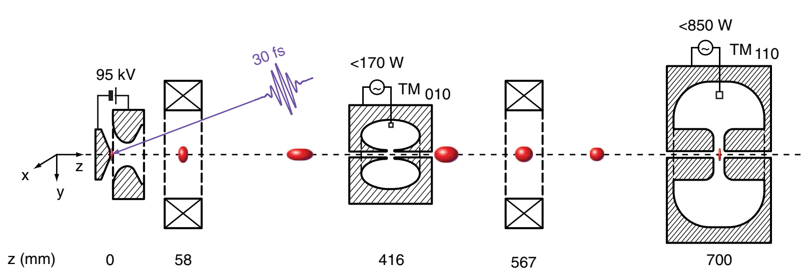

FIG. 3 (color). Schematic of the setup and bunch evolution as it propagates through the beam line.

$$t_b = \mathcal{C}\frac{X_{\rm str}}{B_0(0)}, \qquad (1)$$

with $\mathcal{C} = \gamma m v/(2\pi f e L_{\rm cav} d)$. Here e is the elementary charge, m the electron mass, $\gamma = [1 - (v^2/c^2)]^{-1/2}$ with v the speed of the electrons and c the speed of light, d the distance from the exit of the streak cavity to the MCP, and $L_{\rm cav} = \int_{-\infty}^{\infty} B_0(z)/B_0(0)\cos(2\pi f z/v_z)dz$ the effective cavity length. For our setup $\mathcal{C} = (0.90 \pm 0.01)10^{-11}$ s T m^{-1}. As an independent check we measured the position $x_{\rm scr}$ of the center of the streak on the screen as a function of the rf phase offset, which is equivalent to a change in arrival time. The results for various values of $B_0(0)$ are shown in Fig. 4. A linear fit yields $\mathcal{C} = (1.06 \pm 0.07)10^{-11}$ s T m^{-1}, in satisfactory agreement with the calculated value.

The distribution in the x direction of the streak on the MCP is in fact a convolution of the transverse and longitudinal bunch profile. So, the rms width of the intensity profile on the MCP is given by $\sigma_{\rm MCP} = \sqrt{\sigma_x^2 + X_{\rm str}^2}$, where σ_x is the rms size of the bunch in the x direction at the streak cavity. To increase the temporal resolution we reduce σ_x with a 10 μm slit upstream at a distance of 7 cm from the center of the streak cavity. In Fig. 5 streaks are shown of an uncompressed [Fig. 5(a)] and a maximally compressed [Fig. 5(b)] 250 fC bunch, which have been sliced by the 10 μm slit [Fig. 5(c)] before entering the streak cavity. The solenoid settings are the same for all three images. The horizontal line in [Fig. 5(a)] is due to a local narrowing in the slit. The streak of the uncompressed bunch has the expected ellipsoidal shape with a slight asymmetry in the longitudinal direction, which is caused by image charge forces at the cathode [12]. The streak of the compressed bunch and the image of the slit are slightly tilted, which is due to rotation of the bunch in the tail field of the second solenoid. Both the tilt and the size in the y direction of the maximally compressed bunch [Fig. 5(b)] are slightly different from the projection of the slit [Fig. 5(c)]. This is caused by the compression cavity, which slightly changes both the beam divergence and the energy [10].

The width $\sigma_{\rm MCP}$ is determined by first integrating the sum of five images in the y direction to increase the signal-to-noise ratio, and subsequently fitting the resulting profile to a Gaussian. For streak lengths $X_{\rm str} \approx \sigma_x$ it is necessary to correct for the tilt of the streak: single images are analyzed by taking lineouts through the streak. Each lineout is fitted to a Gaussian and shifted such that all lineouts are centered at the same position. These shifted lineouts are summed and the result is fitted to a Gaussian to obtain $\sigma_{\rm MCP}$.

Figure 1 shows the rms bunch duration t_b as a function of $E_0(0)$. We are able to compress 250 fC bunches from 10 ps down to 100 fs durations. Also shown are the results

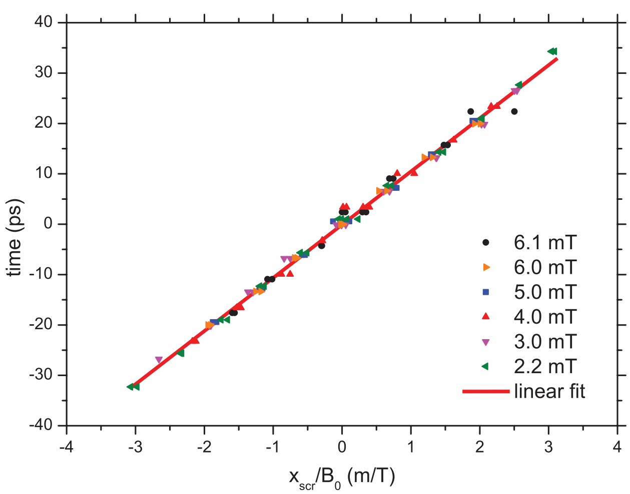

FIG. 4 (color). Time delay as a function of $x_{\rm scr}/B_0$. The red line is a linear fit to the data (black dots). The thickness of the red line reflects the confidence bounds of the fit.

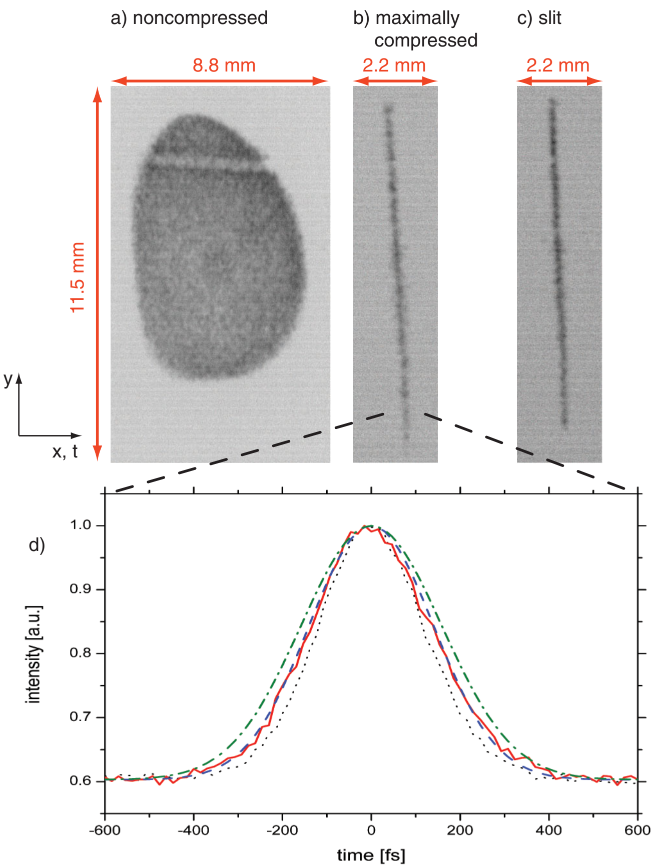

FIG. 5 (color). Streaks of (a) an uncompressed and (b) a maximally compressed 250 fC bunch; (c) projection of the 10 μm slit when the streak cavity is off; (d) streak profiles of a 67 fs bunch (red line), of the projection of the slit (black dotted line), of a Gaussian with $\sigma_{67} = \sqrt{(\sigma_{t,\rm slit})^2 + (67\ {\rm fs})^2}$ (blue dashed line), and of a Gaussian with $\sigma_{100} = \sqrt{(\sigma_{t,\rm slit})^2 + (100\ {\rm fs})^2}$ (green dash-dotted line).

of detailed [15] particle tracking simulations, both with and without the 10 μm slit. The simulations indicate that the presence of the slit does not affect the achievable bunch length appreciably. Clearly the simulations agree quite well with the measurements. Remaining discrepancies are attributed to uncertainties in the charge, laser spot size, and solenoid field strengths. The longitudinal beta function, i.e., the distance in z over which the bunch length increases by a factor of $\sqrt{2}$, cannot be measured directly in our setup. It should, however, be at least as large as the effective cavity length $L_{\text{cav}} = 10.5$ mm, which is in agreement with GPT simulations [10].

At the field strength of maximal compression there is hardly any difference between the streak and the slit projection. Figure 5(d) shows the streak profile of a maximally compressed bunch, the profile of the slit projection, and a Gaussian with $\sigma_{67} = \sqrt{(\sigma_{t,\text{slit}})^2 + (67\ \text{fs})^2}$, where $\sigma_{t,\text{slit}}$ is the slit width converted to the time scale as if it were a streak. The σ_{67} Gaussian fits the measured streak profile quite well, implying a bunch duration $t_b = 67$ fs. The Gaussian with $\sigma_{100} = \sqrt{(\sigma_{t,\text{slit}})^2 + (100\ \text{fs})^2}$ clearly shows that rms bunch durations well below 100 fs have been achieved.

For pump-probe UED experiments the arrival time jitter of the electron bunch with respect to the pump pulse (generally derived from the photoemission laser) is crucial. In our setup phase jitter of the rf compression cavity leads to changes in the bunch velocity, and thus to arrival time jitter at the sample, which can be determined from the measurements shown in Fig. 4. The thickness of the fitted curve reflects the confidence bounds, yielding a rms jitter of 106 fs with respect to the photoemission laser. This arrival time jitter can be translated back into 28 fs phase jitter of the rf field in the compression cavity, which agrees with the expectation based on the 20 fs synchronization accuracy between our 3 GHz oscillator and Ti:sapphire oscillator [16]. The measurements in Fig. 4 have been performed at $E_0 = 2.9$ MV/m. As the arrival time jitter scales linearly with $E_0(0)$ the rms arrival time jitter is 80 fs at the field strength of maximum compression, i.e., $E_0(0) = 2.2$ MV/m.

To show that our bunches have sufficient charge and are of sufficient quality for single-shot UED we carried out a diffraction experiment. We replaced the streak cavity by a standard calibration sample for transmission electron microscopy [17], consisting of a 300 μm copper mesh, a carbon interlayer, and a polycrystalline gold layer of (9 ± 1) nm thickness. A third solenoid is positioned behind the sample with the MCP in its the focal plane. Figure 2 shows a diffraction pattern recorded with a single 0.2 pC, $\sigma_x = (400 \pm 20)$ μm electron bunch, corresponding to 2.5 electrons/μm^2 at the foil. In comparison, Sciaini *et al.* achieved 0.5 electrons/μm^2 at $\gtrsim 350$ fs temporal resolution [6]. Figure 2 also shows the azimuthal integral of the Debye-Scherrer rings. The background due to the grid and the carbon layer has been subtracted from this curve, confirming that the rings are due to diffraction of electrons on the gold film. The curve is fitted using kinematical diffraction theory, with the inelastic scattering cross section and the peak width as fit parameters. The relative positions of the Bragg peaks and their relative intensities are fixed at the theoretical values.

In conclusion, we have demonstrated rf compression of nonrelativistic, space-charge-dominated electron bunches to sub-100 fs durations. Detailed charged particle simulations with the GPT code are consistent with our measurements. Our bunches are suitable for single-shot UED experiments, as we have shown by capturing a high-quality diffraction pattern from a polycrystalline gold film using a single electron bunch.

We are grateful to B. J. Siwick for originally introducing us into the field of UED and many stimulating discussions. We thank E. H. Rietman, A. H. Kemper, and H. A. van Doorn for their expert technical assistance. This research is financially supported by the ''Stichting voor Fundamenteel Onderzoek der Materie'' (FOM), and the Dutch Technology Foundation STW.

*O.J.Luiten@tue.nl

[1] J. R. Dwyer *et al.*, Phil. Trans. R. Soc. A **364**, 741 (2006).
[2] R. Srinivasan *et al.*, Helv. Chim. Acta **86**, 1761 (2003).
[3] P. Emma *et al.*, Nat. Photon. **4**, 641 (2010).
[4] D. A. Shapiro *et al.*, J. Synchrotron Radiat. **15**, 593 (2008).
[5] B. J. Siwick *et al.*, Science **302**, 1382 (2003).
[6] G. Sciaini *et al.*, Nature (London) **458**, 56 (2009).
[7] N. Gedik *et al.*, Science **316**, 425 (2007).
[8] C.-Y. Ruan *et al.*, Science **304**, 80 (2004).
[9] P. Musumeci *et al.*, Rev. Sci. Instrum. **81**, 013306 (2010).
[10] T. van Oudheusden *et al.*, J. Appl. Phys. **102**, 093501 (2007).
[11] O. J. Luiten *et al.*, Phys. Rev. Lett. **93**, 094802 (2004).
[12] P. Musumeci *et al.*, Phys. Rev. Lett. **100**, 244801 (2008).
[13] J. T. Moody *et al.*, Phys. Rev. ST Accel. Beams **12**, 070704 (2009).
[14] T. van Oudheusden *et al.*, in *Ultrafast Phenomena XVI*, edited by P. Corkum, S. De Silvestri, K. A. Nelson, E. Riedle, and R. W. Schoenlein (Springer-Verlag, Berlin, Heidelberg, 2009), p. 938.
[15] http://www.pulsar.nl/gpt
[16] F. B. Kiewiet *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. A **484**, 619 (2002).
[17] Agar Scientific, Cross Grating S106, http://www.agarscientific.com