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Electron source concept for single-shot sub-100 fs electron diffraction in the 100 keV range

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We present a method for producing sub-100 fs electron bunches that are suitable for single-shot ultrafast electron diffraction experiments in the 100 keV energy range. A combination of analytical estimates and state-of-the-art particle tracking simulations show that it is possible to create 100 keV, 0.1 pC, 30 fs electron bunches with a spot size smaller than 500 μm and a transverse coherence length of 3 nm, using established technologies in a table-top setup. The system operates in the space-charge dominated regime to produce energy-correlated bunches that are recompressed by radio-frequency techniques. With this approach we overcome the Coulomb expansion of the bunch, providing a single-shot, ultrafast electron diffraction source concept.
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I. INTRODUCTION

The development of a general experimental method for the determination of nonequilibrium structures at the atomic level and femtosecond time scale would provide an extraordinary new window on the microscopic world. Such a method opens up the possibility of making "molecular movies," which show the sequence of atomic configurations between reactant and product during bond-making and bond-breaking events. The observation of such transition states structures has been called one of the holy grails of chemistry, but is equally important for biology and condensed matter physics.[1–3]

There are two promising approaches for complete structural characterization on short time scales: ultrafast x-ray diffraction and ultrafast electron diffraction (UED). These methods use a stroboscopic—but so far multishot—approach that can capture the atomic structure of matter at an instant in time. Typically, dynamics are initiated with an ultrashort (pump) light pulse and then—at various delay times—the sample is probed in transmission or reflection with an ultrashort electron[4,5] or x-ray pulse.[6] By recording diffraction patterns as a function of the pump–probe delay it is possible to follow various aspects of the real-space atomic configuration of the sample as it evolves. Time resolution is fundamentally limited by the x-ray/electron pulse duration, while structural sensitivity depends on source properties like the beam brightness and the nature of the samples.

Electron diffraction has some unique advantages compared with x-ray techniques:[7] (1) UED experiments are table-top scale; (2) the energy deposited per elastic scattering event is approximately 1000 times lower compared to 1.5 Å x-rays; and (3) for most samples the scattering length of electrons better matches the optical penetration depth of the pump laser. However, until recently femtosecond electron diffraction experiments had been considered unlikely. It was thought that the strong Coulombic repulsion (space-charge) present inside of high-charge-density electron bunches produced through photoemission with femtosecond lasers fundamentally limited this technique to picosecond time scales and longer. Several recent developments, however, have resulted in a change of outlook. Three approaches to circumvent the space-charge problem have been attempted by several groups. The traditional way is to accelerate the bunch to relativistic energies to effectively damp the Coulombic repulsion. Bunches of several hundred femtosecond duration containing high charges (several picocoulombs) are routinely available from radio-frequency (rf) photoguns. The application of such a device in an electron diffraction experiment was recently demonstrated.[8] This is an exciting development; however, energies in the mega-electron volt range pose their own difficulties, including the very short De Broglie wavelength λ ($\lambda \approx 0.002$ Å at 5 MeV), radiation damage to samples, reduced cross section for elastic scattering, nonstandard detectors and general expense of the technology. Due to these and other considerations, electron crystallographers prefer to work in the 100–300 keV range.

A second avenue to avoid the space-charge expansion is by reducing the charge of a bunch to approximately one electron, while increasing the repetition frequency to several megahertz.[9] The temporal resolution is then determined by the jitter in the arrival time of the individual electrons at the sample. According to Ref. 10, simulations show that by minimizing the jitter of the rf acceleration field the individual electrons could arrive at the sample within a time-window of several femtoseconds (possibly even subfemtoseconds). This

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technique, however, requires that the sample be reproducibly pumped and probed $\sim 10^6$ times to obtain diffraction patterns of sufficient quality.

Third, compact electron sources have been engineered to operate in a regime where space-charge broadening of the electron bunch is limited. The current state-of-the-art compact electron gun provides ~300 fs electron bunches, containing several thousand electrons per bunch at sub-100 keV energies and with a beam divergence in the milliradian range.[11,12] This source represents a considerable technical achievement, but is still limited by space-charge effects which limit the number of electrons to less than 10 000 per bunch for applications requiring high temporal resolution. However, because of the relatively low number of electrons per bunch this source does not provide the possibility to operate in single-shot mode.

The ideal source for single-shot transmission UED experiments would operate at (several) 100 keV energies, providing bunches shorter than 100 fs, containing $\gtrsim 10^6$ electrons. The transverse coherence length L_c should be at least a few nanometers—or several unit cell dimension—to ensure high-quality diffraction data. None of the electron source concepts presently in use is able to *combine* these bunch requirements. Herein we present an electron source concept for UED experiments, based on linear space-charge expansion of the electron bunch[7,13] and rf compression strategies,[14] that is able to obtain the ideal parameters presented above with potential well beyond these numbers.

The purpose of this article is twofold. (1) To show on the basis of fundamental beam dynamics arguments and analytical estimations that single-shot, sub-100 fs UED in the 100 keV energy range is *in principle* possible. (2) To show that these conditions can be realized in practive through state-of-the-art particle tracking simulations of a novel, realistic setup.

The remainder of this article is organized as follows. In Sec. II we discuss the beam dynamics of single-shot UED and show that the bunch requirements for single-shot UED can only be reached by operating close to fundamental space-charge limits. The high space-charge density inevitably leads to a fast Coulomb expansion, which needs to be reversed both in the longitudinal and the transverse direction. This can be accomplished with ellipsoidal bunches.[13] In particular we show how the longitudinal expansion can be reversed using the time-dependent electric field of a cylindrical rf cavity resonating in the TM_{010} mode. The beam dynamics discussion and analytical estimates very naturally lead to a setup, which is described in Sec. III. The diode structure of the accelerator, and the rf cavity for bunch compression are described in some detail. Then, in Sec. IV we present the results of our particle tracking simulations, which confirm the analytical estimates and which convincingly show that single-shot, sub-100 fs electron diffraction at 100 keV is feasible. In Sec. V the stability of the setup is discussed. Finally, in Sec. VI, we draw our conclusions.

II. SINGLE-SHOT UED BEAM DYNAMICS

A. General considerations

The transverse coherence length L_c is an important beam parameter in electron diffraction experiments. It is defined as follows in terms of the De Broglie wavelength λ and root-mean-square (rms) angular spread σ_θ:

$$L_c \equiv \frac{\lambda}{2\pi\sigma_\theta}. \tag{1}$$

However, a more general figure of merit of the transverse beam quality, familiar to electron beam physicists, is expressed in terms of the transverse normalized emittance $\varepsilon_{n,x}$, which is defined by

$$\varepsilon_{n,x} \equiv \frac{1}{mc}\sqrt{\langle x^2\rangle\langle p_x^2\rangle - \langle xp_x\rangle^2}, \tag{2}$$

where m is the electron mass, c is the speed of light, x is the transverse position, and p_x is the transverse momentum of an electron. The angular brackets $\langle\,\rangle$ indicate an average over the ensemble of electrons in the bunch. The transverse emittance in the y-direction and the longitudinal emittance in the z-direction are defined analogously. The product of these three emittances is a measure for the phase-space volume occupied by the bunch. Assuming that motions in the x-, y-, and z-directions are decoupled, which is generally a reasonable assumption for freely propagating particle beams, Liouville's theorem states that the emittances are conserved beam quantities. In a beam waist Eq. (2) reduces to $\varepsilon_{n,x} = (1/mc)\sigma_x\sigma_{p_x}$, where σ_x is the rms bunch radius, and σ_{p_x} the rms transverse momentum spread. The transverse coherence length at a beam waist, in particular in a beam focus, is therefore given by

$$L_c = \frac{\hbar}{mc}\frac{\sigma_x}{\varepsilon_{n,x}}, \tag{3}$$

where $\hbar$ is Planck's constant. When aiming for $L_c \geq 4$ nm and $\sigma_x \leq 0.2$ mm at the sample placed in a beam focus, then it necessarily follows from Eq. (3) that $\varepsilon_{n,x} \leq 0.02$ mm mrad. The precise requirements on the transverse coherence length to obtain a diffraction pattern of sufficient quality will however depend on the sample. Moreover, recording a diffraction pattern in a single shot requires a bunch charge of at least 0.1 pC. Such low-emittance, highly charged, ultrashort bunches can only be created by pulsed photoemission.[15] The initial transverse emittance for pulsed photoemission from metal cathodes is $\varepsilon_{i,x} = 8 \times 10^{-4}\sigma_x$,[15] so that the initial rms radius σ_x at the photocathode may not be larger than 25 μm. Extracting a charge $Q = 0.1$ pC in an ultrashort pulse from such a small spot leads to an image charge density $Q/2\pi\sigma_x^2$ and therefore to an image-charge field $E_{\text{image}} = Q/4\pi\varepsilon_0\sigma_x^2 \approx 1$ MV/m,[16] where ε_0 is the permittivity of vacuum. Acceleration of the bunch requires the acceleration field to be substantially higher, i.e., about 10 MV/m.

The space-charge fields inside the bunch are of the same order of magnitude as the image charge fields, resulting in a

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rapid expansion of the bunch to millimeter sizes within a nanosecond, as will be shown in the next section.

Up to now such a space-charge explosion was considered unavoidable, and strategies were developed aiming at minimizing its effect either by setting an upper limit to the charge of a bunch[7,17] or by accelerating the bunch to relativistic velocities.[8] In this article, however, we show that the space-charge expansion is not necessarily a problem, provided that the expansion results in a bunch with a linear velocity-position correlation.

B. Expansion and compression of ellipsoidal bunches

To be able to compress an electron bunch, both transversely *and* longitudinally, to the required dimensions while conserving its emittance, it is necessary that the rapid space-charge induced expansion is reversible; i.e., the space-charge fields must be linear, which is precisely the case for a homogeneously charged ellipsoidal bunch.[18] Such a bunch can actually be created in practice by femtosecond photoemission with a "half-circle" radial laser profile.[13] The expansion in the transverse direction can be reversed by regular charged-particle optics, such as magnetic solenoid lenses. The reversal of the expansion in the longitudinal direction, i.e. bunch compression, is less straightforward. Several methods have been developed for relativistic accelerators, employing either constant magnetic fields[19] or time-dependent electric fields.[14]

In this article we propose to use the time-dependent axial electric field of a cylindrical 3 GHz rf cavity oscillating in the TM_{010} mode. The idea is to apply a ramped electric field, such that the front particles, which move the fastest, are decelerated while the slower electrons at the rear of the bunch are accelerated, leading to ballistic compression in the subsequent drift space. The field ramp needs to be timed very accurately, as it has to coincide with the picosecond bunch. This can be realized by using a rf field, whose phase can be synchronized to the femtosecond photoemission laser pulse with an accuracy better than 50 fs.[20]

We start by looking into the expansion dynamics of an ellipsoidal bunch, which involves the conversion of electrostatic potential energy into kinetic energy. Inside a uniformly filled spheroid (a cylindrically symmetric ellipsoid) with maximum radius R, and half-length L the electrostatic potential $V(r,z)$ as a function of the radial coordinate $r=\sqrt{x^2+y^2}$ and the longitudinal coordinate z is given by[21]

$$V(r,z) = \frac{\rho_0}{2\varepsilon_0}(MR^2 - M_r r^2 - M_z z^2), \tag{4}$$

where $\rho_0=3Q/4\pi R^2 L$ is the charge density, $M=\arctan(\Gamma)/\Gamma$, $M_r=\frac{1}{2}(1-M_z)$, $M_z=[(1+\Gamma^2)/\Gamma^3][\Gamma-\arctan(\Gamma)]$, with the eccentricity of a spheroid $\Gamma=\sqrt{R^2/L^2-1}$.[22] The potential given by Eq. (4) is defined such that it equals zero if $\rho_0=0$, i.e., if $R, L\to\infty$. Using $\mathbf{E}=-\nabla V$ it follows immediately that the space-charge electric fields are indeed linear functions of position:

$$E_r(r) = \frac{\rho_0}{\varepsilon_0}M_r r, \tag{5}$$

$$E_z(z) = \frac{\rho_0}{\varepsilon_0}M_z z. \tag{6}$$

The linear space-charge fields give rise to particle velocities which are also linear functions of position. The space-charge fields of a uniform ellipsoidal bunch thus lead to a linear expansion, with the result that the uniform ellipsoidal—and thus linear—character of the bunch is maintained. In our approach we initiate the bunch by pulsed photoemission with a femtosecond laser pulse with a half-circle transverse intensity profile $I(r)=\sqrt{1-(r/R)^2}$. As shown in Ref. 13 this is essentially equivalent to starting out with a flat, pancake-like spheroid $(L \lll R)$. During the subsequent acceleration this pancake bunch automatically evolves into a three-dimensional, hard-edged uniform spheroid. During this evolution the potential energy of the pancake bunch is converted into kinetic energy of the electrons. In the limit for a disk of zero thickness, i.e., $L\to 0$, the potential energy of a uniform ellipsoidal bunch remains finite and is given by (see Appendix A)

$$U_{p,\mathrm{disk}} = \frac{3Q^2}{40\varepsilon_0 R}. \tag{7}$$

For a uniform ellipsoidal bunch with linear velocity-position correlations $\mathbf{v}(r,z)=(r/R)v_t\mathbf{e}_r+(z/L)v_l\mathbf{e}_z$ the total kinetic energy is given by (see Appendix A)

$$U_k = \frac{N}{5}mv_t^2 + \frac{N}{10}mv_l^2, \tag{8}$$

where $N=Q/e$ is the number of electrons in the bunch. The space-charge-induced expansion of the bunch ends up in a ballistic expansion with an asymptotic velocity that can be calculated with Eqs. (7) and (8). Assuming that the longitudinal and transverse asymptotic velocities are equal,[23] i.e., $v_l=v_t$, the electrons at the extremities of the bunch reach an asymptotic velocity $v_l=(Qe/4m\varepsilon_0 R)^{1/2}$. For a 0.1 pC bunch of 50 μm radius this results in an asymptotic velocity $v_l=3.2\times10^6$ m/s.

Interestingly, the value of the space-charge-induced asymptotic velocity difference $2v_l$ is not equal to the final value of $2v_l$ after the bunch has left the acceleration field. Due to the fact that the slower particles at the back spend a longer time in the acceleration field than the particles at the front of the bunch, the slower particles gain additional momentum from the field. In this way the space-charge induced velocity difference $2v_l$ is reduced by the "longitudinal exit kick" of the acceleration field. Suppose the space-charge expansion is completed in a very short time, i.e. the asymptotic velocity difference $2v_l$ is reached after a distance the bunch has traveled much smaller than the acceleration gap. In a uniform acceleration field E_{acc} the bunch duration τ at the end of the diode is then $\tau=2mv_l/eE_{\mathrm{acc}}$, which implies that the particles at the back of the bunch acquire an additional momentum $eE_{\mathrm{acc}}\tau=2mv_l$, canceling the space-charge-induced expansion speed. In reality however, this cancelation is not complete, since we have neglected the finite time it takes to complete the space-charge expansion. But clearly the final velocity difference $2v_l$ is reduced substantially due to the longitudinal exit kick of the diode.

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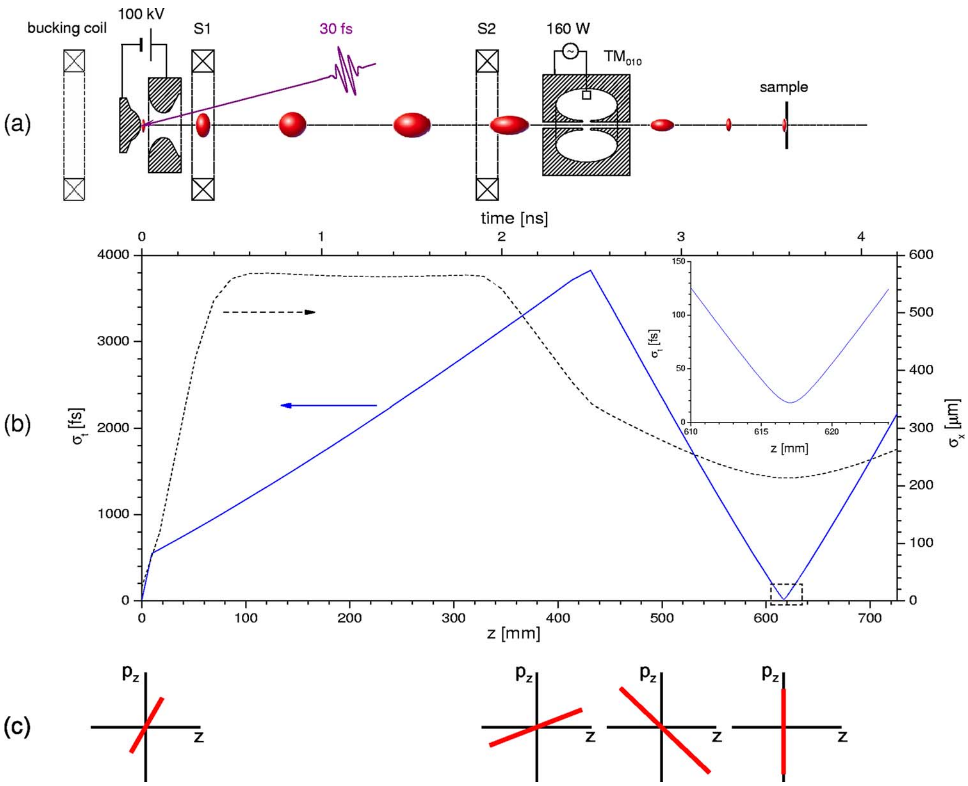

FIG. 1. (Color online) (a) Schematic of the proposed setup. The setup is to scale, the bunches serve only as a guide to the eye. (b) rms bunch duration σ_t (solid line) and rms bunch radius σ_x (dashed line) as a function of position z and time. The inset shows a closeup of σ_t as a function of z around the focus position, which is indicated by the dashed square. (c) Schematics of the longitudinal phase-space distribution of the electron bunch at several key points in the setup.

Now that we have described the expansion dynamics of an ellipsoidal bunch, let us take a look at the compression of the bunch. Suppose the rf cavity has reversed the longitudinal velocity-position correlation, so that it is now given by $v_z = -(z/L)v_l$. Using the same energy conversion considerations as for the expansion case we now estimate the required velocity difference $2v_l$ for ballistic compression of the bunch. Assume that the potential energy of the expanded bunch is much smaller than its kinetic energy, i.e. $U_p \ll U_k$. Further, it is assumed that the beam has been collimated. The bunch thus has a linear velocity-position correlation with the transverse expansion speed much smaller than the longitudinal one: $v_t \ll v_l$. At the time-focus the bunch has reached its shortest possible length and $v_l=0$: all kinetic energy has been converted into potential energy. With Eqs. (7) and (8) it can then be calculated that for an ellipsoidal bunch with charge $Q=0.1$ pC and radius $R \approx 2\sigma_x = 400$ μm the required velocity difference for this ballistic compression is $2v_l = 3.9 \times 10^6$ m/s. In Appendix B we show that a rf cavity can introduce a maximum momentum difference $\Delta p = 2mv_l$ between the most outward electrons of a bunch given by

$$\Delta p = \frac{eE_0\omega\tau d}{v_c}, \tag{9}$$

where $\tau = 2L/v_c$ is the duration of the bunch e is the elementary charge, E_0 is the amplitude, ω is the frequency of the rf field, d is the cavity length, and v_c is the velocity of an electron at the center of the bunch. From Eq. (9) it follows that the longitudinal momentum rf kick that is required for ballistic compression of a 100 keV bunch with duration $\tau = 3$ ps can be realized by a rf field with amplitude $E_0 = 6.5$ MV/m, in a cavity with resonant frequency $f = \omega/2\pi = 3$ GHz and a length $d = 1$ cm.

III. SINGLE-SHOT UED SETUP

A. Overview

As an implementation of the ideas presented in Sec. II, we propose a table-top UED setup as shown in Fig. 1(a), consisting of a dc photogun, two solenoidal magnetic lenses $S1$ and $S2$, and a rf cavity. The bucking coil is to null the magnetic field at the cathode surface. Electrons are liberated from a metal photocathode by a transversely shaped, ultrashort laser pulse and accelerated through a diode structure to an energy of 100 keV. By applying a dc voltage of 100 kV between the cathode and the anode an acceleration field of 10 MV/m is obtained. Because of the linear space-charge fields the photoemitted bunch evolves such that its phase-space distribution becomes linearly chirped with faster electrons toward the front and slower electrons toward the back. This is indicated in Fig. 1(c) by the schematic longitudinal phase-space distribution, i.e. longitudinal momentum p_z versus position z in the bunch. The electric field oscillating in the TM_{010} mode in the rf cavity either accelerates or decelerates electrons passing through along the axis, depending on the rf phase. By injecting a bunch just before the field goes through zero, the front electrons are decelerated and the back electrons are accelerated. In this way the velocity-correlation in the bunch is reversed. To illustrate this scheme Fig. 1(c) shows the longitudinal phase-space distribution of the bunch at several key points in the setup.

B. dc photogun design

We have designed a 100 kV dc photogun with the SUPERFISH code.[24] A bulk copper cathode is used, without a grid in front of it. Instead, an anode is used with a circular hole in it with a radius much larger than the typical beam

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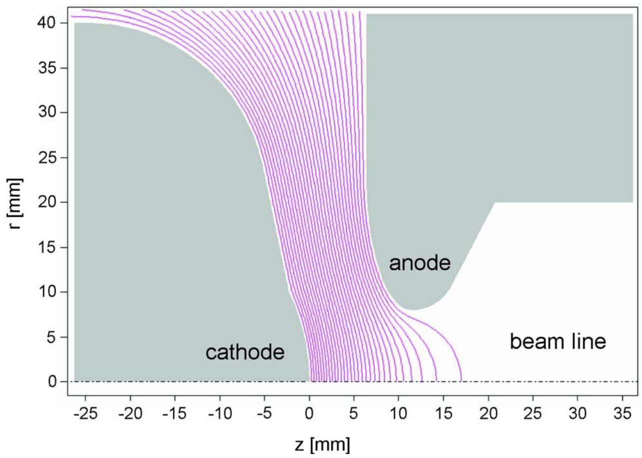

FIG. 2. (Color online) The SUPERFISH design of the diode structure of the dc photogun. The dash-dotted line is the axis of rotational symmetry. The purple lines are equipotential lines.

radius. In this way field nonlinearities, which could lead to irreversible emittance growth, are minimized. The shapes of the cathode and the anode have been designed such that the highest field strength of 116 kV/cm is at the center of the cathode, while minimizing the divergence of the field around the particle trajectories. The center of the cathode is a flat circular area with a diameter of 1 mm, which is much larger than the laser spot size. The diode geometry is shown in Fig. 2.

C. rf cavity design

The rf cavity has also been designed with the SUPERFISH code.[24] We have designed an efficient cavity which only requires 420 W input power to obtain the required field strength of 6.5 MV/m (see Sec. II B). This is a power reduction of about 90% compared with the regular pillbox geometry. 3 GHz rf powers up to 1 kW can be delivered by commercially available solid state rf amplifiers, so klystrons are not required. Further, for transportation of the power from the rf source to the cavity coaxial transmission lines can be used instead of waveguides. Energy coupling between the coax line and the cavity can be established with so-called magnetic coupling by bending the inner conductor of the coax into a small loop inside the cavity. The cavity design is shown in Fig. 3.

IV. PARTICLE TRACKING SIMULATIONS

The setup has been designed and optimized with the aid of the General Particle Tracer (GPT) code.[25] The bunch charge of 0.1 pC allows us to model the electrons in the bunch such that each macroparticle represents a single electron.

The electric fields of both the dc accelerator and the rf cavity, as presented in the previous section, have been calculated with the SUPERFISH set of codes[24] with 10 μm precision. The solenoids are modeled by a fourth-order off-axis Taylor expansion from the analytical expression for the on-

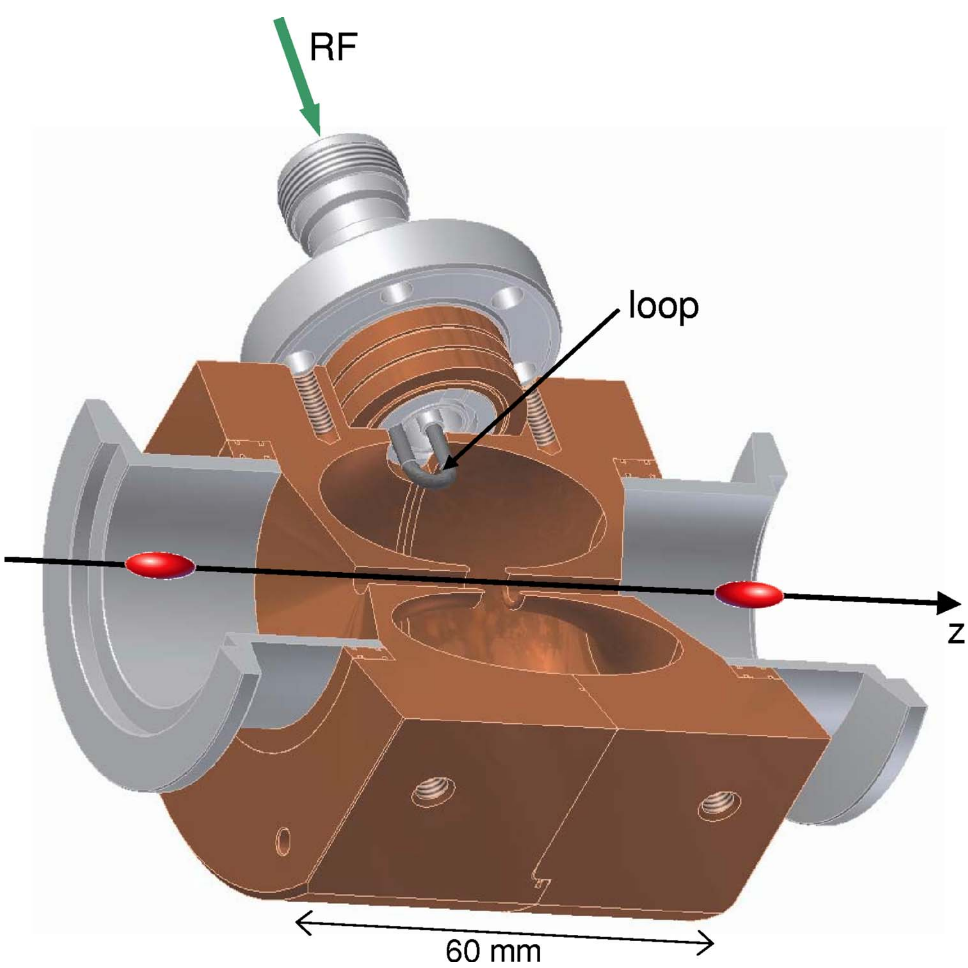

FIG. 3. (Color online) Design of the rf cavity including the loop for energy coupling between the coax line and the cavity. The width of the gap where the electron bunches pass through is 6 mm, the total cavity width is 60 mm. The electron bunches are not to scale and serve only as a guide to the eye.

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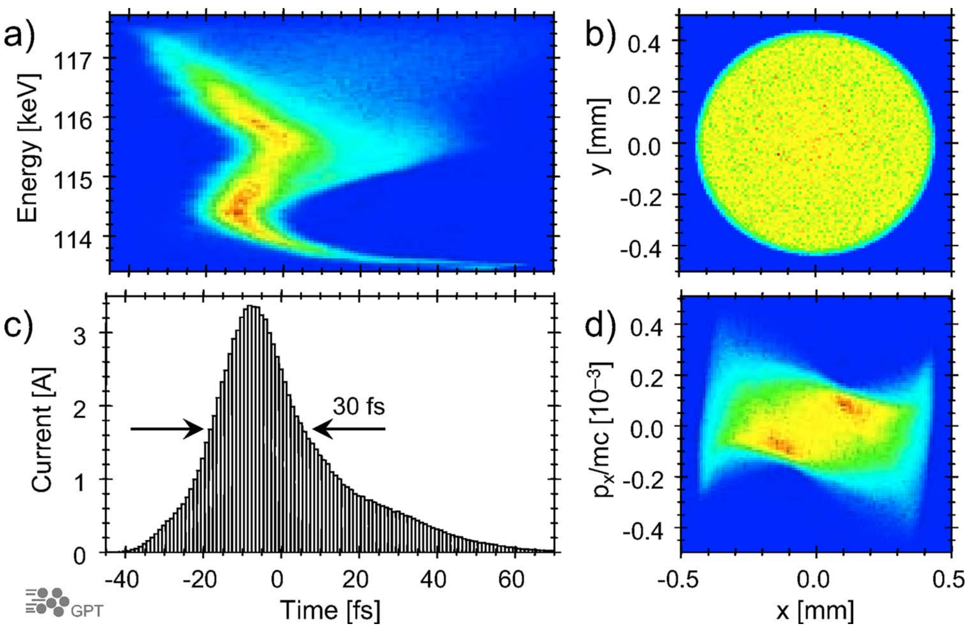

FIG. 4. (Color online) (a) Longitudinal phase-space distribution, (b) cross section, (c) current distribution, and (d) transverse phase-space distribution of the electron bunch at the sample.

axis field. The effect of space charge is accounted for by a particle in cell method based on a three-dimensional anisotropic multigrid Poisson solver, tailor made for bunches with extreme aspect ratios.[26,27] Image charges are taken into account by a Dirichlet boundary condition at the cathode. Wakefields are not taken into account, but because of the low energy of the electrons, the low charge of the bunch, and the low peak current, these fields can be neglected.

The ideal initial half-circle electron density profile is approximated by a Gaussian transverse profile with a standard deviation of $\sigma_x=50$ μm truncated at a radius of 50 μm, corresponding to the one-sigma point. This profile is experimentally much more easy to realize and turns out to be sufficient to produce bunches with the required parameters at the focus. To simulate the photoemission process GPT creates a Gaussian longitudinal charge density profile with a full-width-at-half-maximum (FWHM) duration of 30 fs. An isotropic 0.4 eV initial momentum distribution is used to model the initial emittance.

The optimized position of the rf cavity, at $z=430$ mm, is a trade-off between desired longitudinal space-charge expansion to a few picoseconds before injection and unavoidable accumulation of nonlinear effects. The position and on-axis field strength of solenoid $S2$, 334 mm and 0.03 T respectively, have been chosen such that the beam waist at the sample has the desired size and coincides with the time-focus.

The rf phase of the cavity must be tuned to minimize nonlinear effects in the longitudinal compression. The optimized phase is a slight deceleration: 11° off the zero crossing. To compensate for this slight rf deceleration the voltage of the dc accelerator has been raised from the nominal value of 100 kV to 120 kV to ensure the electron bunch has at least 100 keV kinetic energy at the sample. Solenoid $S1$ is located at $z=50$ mm, and produces an on-axis field of 0.05 T to collimate the beam. The amplitude of the cavity field is $E_0=4$ MV/m, which is lower than the result of the analytical calculation in Sec. II B. However, there it was assumed that the bunch had a constant rms radius $\sigma_x=200$ μm, whereas from Fig. 1(b) it is clear that the radius is almost twice as large when the longitudinal compression starts. This larger radius results in a lower longitudinal space-charge field, so that a smaller compression field strength is required. Moreover, the assumption of a constant radius in the analytical calculation implies that the electrons have no transverse velocity, which is of course not the case: while longitudinal compression takes place the bunch is also transversely compressed. The contribution of the transverse velocity to the initial kinetic energy of the bunch is thus neglected in the analytical calculation.

The bunch evolution in the optimized setup is shown in Fig. 1(b). Due to the high space-charge fields the expansion becomes ballistic quickly after initiation of the bunch. The transverse and longitudinal asymptotic velocities are respectively $v_t=2.9\times10^6$ m/s and $v_l=3.5\times10^6$ m/s. These results are in good agreement with the analytical estimates in Sec. II B. After the diode the transverse beam-size is mainly determined by the two solenoids, but there is also a slightly defocusing effect of the rf cavity. When leaving the diode the longitudinal expansion speed drops abruptly by one order of magnitude to $v_l=0.5\times10^6$ m/s due to the longitudinal exit kick of the diode, as explained in Sec. II B. The bunch then ballistically expands to a several picosecond duration to be recompressed by the rf cavity to below 30 fs. From Fig. 1(b) it follows that this ballistic compression happens with a velocity difference $2v_l=2.4\times10^6$ m/s, which is slightly smaller than the result of the estimation in Sec. II B. According to Eq. (9) this velocity difference can be induced with a rf field strength of only 4 MV/m, which is in perfect agreement with the value of this parameter in the simulation.

Figure 4 shows several projections of the phase-space distribution of the bunch at the sample: (a) the longitudinal phase-space distribution, (b) the transverse cross section, (c) the current distribution, and (d) the transverse phase-space distribution. At the sample the 0.1 pC bunches are characterized by a rms duration $\sigma_t=20$ fs, a rms radius $\sigma_x=0.2$ mm,

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a transverse coherence length $L_c = 3$ nm, an average kinetic energy $U_k = 116$ keV, and a relative rms energy spread $< 1\%$. In addition to the current distribution it is noted that the FWHM bunch duration of 30 fs covers 68% of the electrons in the bunch. A bunch duration of 100 fs covers 99.5% of the electrons. From Fig. 4 it is clear that the setup shown in Fig. 1(a) provides a practical realization of a device capable of producing electron bunches that fulfill *all* the requirements for single-shot UED.

Of all bunch parameters only the bunch duration is strongly dependent on the longitudinal position: over a range of 5 mm around the target position, i.e. $z = (617 \pm 2.5)$ mm, the rms bunch duration varies between 20 and 50 fs, whereas the other parameters do not change significantly. To determine the location of the focal point in practice the bunch length has to be measured, which can be done with e.g. laser ponderomotive scattering[11,28] or Coulomb scattering with an electron cloud that is photoemitted from a metal grid.[2]

V. STABILITY CONSIDERATIONS

For pump–probe experiments the arrival-time jitter should be less than the bunch duration, requiring a voltage stability of 10^{-6} for the power supply of the accelerator. This constraint is also more than sufficient for stable injection on the proper phase of the rf cavity. Such stable voltage supplies are commercially available. A second requirement is that the laser pulse is synchronized to the rf phase, also with an accuracy of less than the bunch duration. We have developed a synchronization system that fulfills this condition.[20] This leaves the initial spot size as the main experimental parameter that influences the bunch quality. Simulations show that a deviation of 10% in spot size decreases the coherence length by 0.2 nm as theoretically expected, while the bunch radius and bunch length at the sample do not change significantly.

VI. CONCLUSIONS

In summary, we have presented a robust femtosecond electron source concept that makes use of space-charge driven expansion to produce the energy-correlated bunches required for radio-frequency compression strategies. This method does not try to circumvent the space-charge problem, but instead takes advantage of space-charge dynamics through transverse shaping of a femtosecond laser pulse to ensure the bunch expands in a reversible way.[13] This reversibility enables six-dimensional phase-space imaging of the electron bunch, with transverse imaging accomplished by regular solenoid lenses and longitudinal imaging by rf bunch compression. Based on fundamental beam dynamics arguments and analytical estimates we have shown that in principle it is possible to create a 100 keV, 0.1 pC, sub-100 fs electron bunch, which has a spot size smaller than 500 μm and a transverse coherence length of several nanometers. The results of our GPT simulations, which are consistent with the analytical estimates, convincingly show it is possible to realize such a bunch in realistic accelerating and focusing electric fields. We have designed a compact setup to create electron bunches that are suitable for single-shot, ultrafast electron diffraction experiments. With these bunches it will be possible for chemists, physicists, and biologists to study atomic level structural dynamics on the sub-100 fs time scale.

APPENDIX A: KINETIC AND POTENTIAL ENERGY OF AN ELLIPSOIDAL BUNCH

The potential energy U_p of a homogeneously charged spheroidal electron bunch is given by

$$U_p = \frac{1}{2}\int \rho_0 V(\mathbf{r}) d^3 r = \pi \int_{-L}^{L} dz \int_0^{R\sqrt{1-z^2/L^2}} \rho_0 V(r,z) r\, dr, \tag{A1}$$

where R and L are the maximum radius and maximum half-length of the bunch, respectively. The charge density is given by $\rho_0 = 3Q/4\pi R^2 L$, with Q the charge of the bunch. With the potential $V(r,z)$ of a uniformly charged ellipsoid, as given by Eq. (4), this is leading to

$$U_p = \frac{3}{20}\frac{Q^2}{\pi\varepsilon_0 L}\frac{\arctan(\Gamma)}{\Gamma}. \tag{A2}$$

The velocity of the particles in a linearly chirped bunch is given by $\mathbf{v}(r,z) = (r/R)v_t \mathbf{e}_r + (z/L)v_l \mathbf{e}_z$. The total kinetic energy of all electrons in an ellipsoidal bunch together is then in the bunch's rest frame given by

$$U_k = \frac{m\rho_0}{2e}\int_{-L}^{L} dz \int_0^{R\sqrt{1-z^2/L^2}} r\, dr \int_0^{2\pi} |\mathbf{v}(r,z)|^2 d\varphi$$

$$= \frac{N}{5} m v_t^2 + \frac{N}{10} m v_l^2. \tag{A3}$$

APPENDIX B: MOMENTUM MODULATION BY A TM_{010} ELECTRIC FIELD

To calculate the momentum difference that a TM_{010} electric field introduces between the most outward electrons of a bunch we first assume that all electrons initially have the same velocity v_c and that the subsequent velocity changes are so small that the resulting changes in the transit times through the rf cavity are negligible. The momentum change Δp_1 of a single electron entering the cavity at time t_1 is then given by

$$\Delta p_1 = -\int_{t_1}^{t_1 + d/v_c} eE(t) dt, \tag{B1}$$

where d is the cavity length, e is the elementary charge, and time is represented by t. The electric field is given by $E(t) = E_0 \sin(\omega t - (\omega d/2v_c) + \phi_0)$, with amplitude E_0, frequency ω, and phase offset ϕ_0 such that if $\phi_0 = 0$ the center electron of the bunch will have no net momentum change after the rf cavity. For an electron at the front of a bunch $t_1 = -L/v_c$, whereas for an electron at the back $t_1 = L/v_c$. The momentum changes of these two electrons can be calculated separately. With the assumptions $\omega L/v_c \ll 1$ and $\omega d/2v_c \ll 1$ subtraction of these momenta is leading to a momentum difference between an electron at the front and an electron at the back of the bunch, given by $\Delta p = (eE_0 \omega\tau d/v_c)\cos(\phi_0)$, where τ

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$=2L/v_c$ is the bunch duration at the moment of injection into the cavity.

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