Radio-frequency compressed electron pulse-width characterization by cross-correlation between electron bunches and laser-induced plasma*

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A method is proposed to determine the temporal width of high-brightness radio-frequency compressed electron pulses based on cross-correlation technique involving electron bunches and laser-induced plasma. The temporal evolution of 2-dimensional transverse profile of ultrafast electron bunches repelled by the formed transient electric field of laser-induced plasma on a silver needle is investigated, and the pulse-width can be obtained by analyzing these time-dependent images. This approach can characterize radio-frequency compressed ultrafast electron bunches with picosecond or sub-picosecond timescale and up to 10^5 electron numbers.

Keywords: radio-frequency compression, electron pulse-width, laser-induced plasma, ultrafast electron diffraction

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1. Introduction

Ultrafast structural dynamics occurring on picosecond or even sub-picosecond timescales induced by femtosecond laser pulses are of great interest in the field of solid-state physics, gas-phase chemical reaction, and biological sciences.[1–4] Time-resolved femtosecond laser spectroscopy is commonly used in these studies,[5,6] but the structural information resolving from the spectrum remains elusive. The recently developed ultrafast electron diffraction (UED) technology becomes a hot point in providing direct imaging of spatial structure changes at an atomic level.[7–9] Measuring structural dynamic processes by UED technology are mostly to use a pump-probe scheme in which one laser pulse is employed as a pump beam and another time-correlated electron pulse is used as a probe beam.[10,11] Then the atomic "structure movie" can be clearly seen by recording the time-dependent diffraction patterns. The pulse-width and brightness of the ultrafast electron pulses are important for the UED system, which determines its temporal resolution and detection sensitivity.

The electron pulse originally copies the pulse-width of the driven laser, but it will be largely broadened during propagation due to the space charge (SC) effect, which results from the electron exclusion and becomes stronger with the electron brightness increasing.[12] For example, the pulse-width of electron bunch containing 10^5 electrons will reach more than 20 ps after 5-ns propagation according to the mean-field simulation model.[13] Traditionally, the decrease of the electron number can effectively reduce the SC effect, but it will result in insufficient brightness and low sensitivity. The recently proposed direct-current (DC) extraction and radio-frequency (RF) compression technique shows large potential in generating 0.1-pC sub-picosecond electron pulses with the energy in a range of 40 keV–100 keV, which takes advantage of space-charge dynamics instead of trying to circumvent the electron expansion.[14] In this apparatus, the electron bunches emitted from the back-irradiated cathode are first accelerated by a 10-MV/m DC electric field and subsequently pass through an RF compression cavity to acquire a velocity chirp which is exactly opposite to the SC expansion, and it will be compressed at a further propagation distance depending on the RF field phase and amplitude.

The measurement of high-brightness electron pulse-width is a great experimental challenge in RF-compressed UED system. Recently, Kassier *et al.* suggested a new streak camera technology with mm-long streaking plates and ultralow timing jitter for the measurement of bright pulses in ultrafast electron diffraction.[15] Gao *et al.* demonstrated that the ponderomotive scattering between the laser field and electrons can be employed to characterize RF compressed electron pulses.[16] In this paper, we suggest a very simple way of characterizing high-brightness RF compressed electron pulses based on electron–plasma cross-correlation technique,[17,18] which is commonly used in the UED community to determine the approximate time-zero position.[19] This method can measure the electron pulse-width without employing more devices while determining the zero-time position. The tempo-

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ral resolution is 1.3 ps, which can be used in ultrafast structural dynamics in which needed are the high-brightness electron pulses on a picosecond time scale, such as martensitic phase transformation process of iron,[20] nucleation and crystallization phenomena.[21]

2. Basic principle

The suggested method for electron pulse-width determination is based on the interaction between the electrons and the transient electric field of laser-induced plasma on a silver needle in high vacuum. With intense femtosecond laser excitation, most of low energy electrons will be ejected from the silver surface through multi-photon photoemission and thermionic emission.[19] They accumulate in the photo-excited surface area and produce a strong transient electric field, which grows and reaches its maximum and then fades out gradually. Though the whole process last more than a few hundreds of picoseconds, the buildup time (from zero to its maximum) persists just for several picoseconds.[19]

The passing electrons will be repelled by laser-induced plasma field, which can be seen from the temporal evolution of two-dimensional (2D) transverse profile of ultrafast electron bunches. The excluded electron number depends on the strength of laser-induced plasma field and the electron temporal distribution in each single pulse, and it achieves its maximum when the peak of the laser-induced plasma field is synchronized with the center electrons of each pulse. The corresponding rise-time can be obtained by analyzing the repelling process with different time delays. The rise-time reflects the dynamics of the laser-induced plasma field buildup and its interaction with the electron beam, of which the value is mainly determined by the buildup time of the plasma field, the laser and electron pulse-width and the temporal mismatch resulting from the velocity difference between the two.[17] Commonly, the laser pulse-width is on a femtosecond scale and can be neglected, while the buildup time and the temporal mismatch are on a picosecond scale which set the measurement limit.

3. Experimental setup

Figure 1 shows a home-built UED setup. The system is composed of four parts, i.e., a femtosecond regenerative amplification laser system (Spitfire Ace Ultrafast Amplifiers-35f, Spectra-Physics), a home-built laser driven electron gun, a sample transfer system (Nor-Cal), and an electron detection system. The laser pulse with a central wavelength of 800 nm and a pulse duration of 50 fs is divided into two paths by a beamspliter (BS), one (70%) is focused on the silver tip to generate plasma, and the other (30%) is coupled into the electron gun to generate the photoelectron bunches after the frequency has been tripled by a third-harmonic generation (THG) setup. To obtain a high conversion efficiency of the THG, a collinear configuration consisting of three collinear β-barium borate crystals (BBO) is employed.[22] The remaining fundamental light is filtered out by a 266-nm optical filter. The produced electron bunches are accelerated by a 10-MV/m electric field to 40 keV$\sim$50 keV energy and reshaped by a 100-μm pinhole on the anode. Then the electron pulses are transversely focused by a magnetic lens and longitudinally compressed by a 3.2-GHz RF cavity with the TM_{010} mode which is driven by an RF generator and phase-locked to a femtosecond laser oscillator. The electron bunches can be adjusted by two pairs of deflection plates. The electron images are multiplied

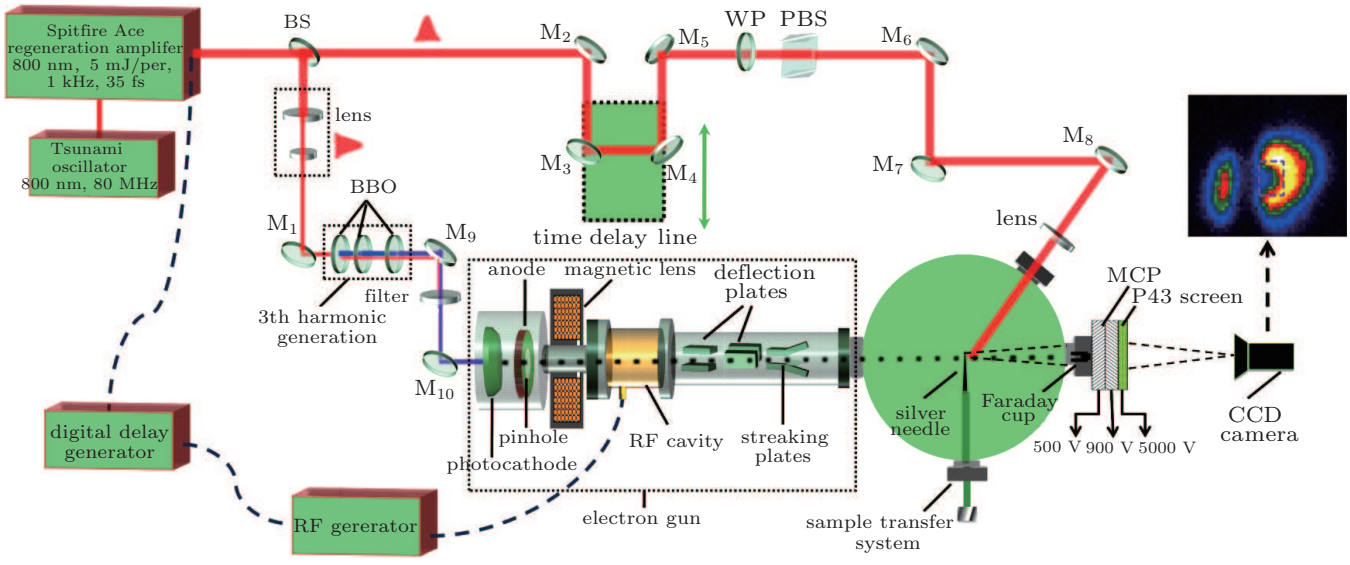


Fig. 1. (color online) Schematic diagram of the UED setup and the geometry of cross-correlation experiment. The output 800-nm lasers are indicated by red solid lines and 266 nm by purple thin solid lines. The electric signals are denoted with blue dashed lines.

by a pair of microchannel plates (MCP, Photek) coupled to a P43 phosphor screen and recorded by a charge coupled device camera (CCD, DU888 Andor). A Faraday cup is equipped to measure the number of electrons per pulse in front of the detection system. The high purity silver needle is mounted on the end of sample transfer system. The experiment is arranged with a pump-probe configuration, and the time delay between the femtosecond laser and electron pulses can be accurately controlled by a motor-driven linear delay line with fs precision. The power of the pump laser pulses can be continuously attenuated by a half-wave plate (WP) and a polarized beam-spliter cube (PBS), and all the timing sequence is controlled by a digital delay generator (DG645, Stanford Research System).

4. Result and discussion

The evolution of 2D electron bunch transverse profile with delay time is displayed in Fig. 2, where each image is captured with 0.2-s exposure time containing 200 shots, and the number of electrons per pulse is measured to be 3.5×10^4 by the Faraday cup. It can be clearly observed that the electron bunches are divided into two regions by the silver needle. The time zero point ($T = 0$ ps) can be defined as the onset change of the electron profile because the laser-induced plasma and the accompanying electric field are immediately built after excitation.[17] After T= 0 ps, a semicircular shadow appears and gradually expands along the direction perpendicular to the needle, which indicates that the electron bunches are distorted by the laser-induced plasma field. The green fluorescence on the needle surface in the same direction could be clearly seen from the observation window equipped on UED setup shown in Fig. 1. This demonstrates that the laser-induced plasma and the repulsive electric field around the silver target are generated and accumulated in this direction, scattering the passing electron bunches. The size of the semicircular shadow achieves its maximum at 25 ps and then diminishes, while the intensities of the shadow pixels increase and return to the original values after about one hundred ps.

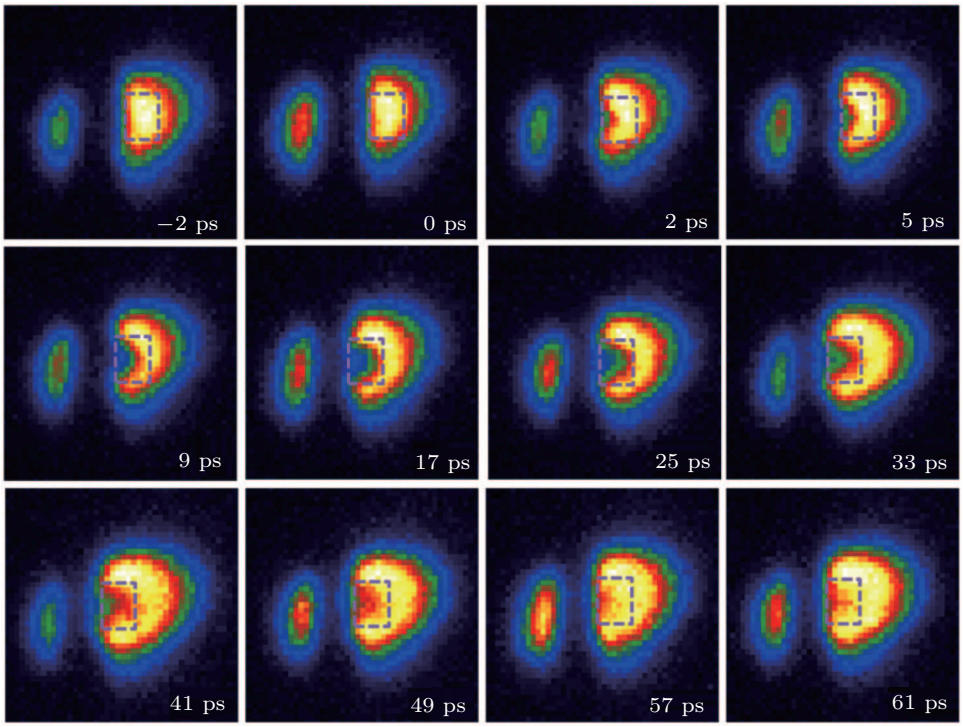


Fig. 2. (color online) The electron bunch transverse profile evolution with delay time, caused by the laser-induced plasmas on the silver needle. The time delay between the pump laser and probe electron pulses is shown on the bottom right corner. The dotted box is marked as the selected region for further data analysis.

To quantitatively analyze the interaction between the electron bunches and laser-induced plasma, the pixel intensity is integrated over the shadow area. The region selected for data analysis is the same at each delay time, which is determined as the maximum area of electron beam repelled by the plasma electric field. Figure 3 shows the dependences of normalized intensity on time delay at different laser intensities, here the experimental data are normalized by the electron intensity at the negative time delay. The turning point of the two fitting lines can be defined as the zero time position between the pump laser and probe electron pulses. It is found that the intensity decrease is strongly dependent on excitation laser in-

tensity, while the decay time is nearly constant. For example, the decrease of intensity at a laser intensity of 40 mW is 20% larger than that measured at 10 mW, but the decay times are fitted to be nearly identical (11.8 ps and 12.0 ps, respectively) by a single exponential function. The decay time is defined as the time constant τ, from which the pulse-width of high-brightness RF compressed electron bunches can be derived. The area selected for further analysis introduces some uncertainty, which only affects the intensity decrease and does not change the time constant, that is, it shows no influence on our measurement results.

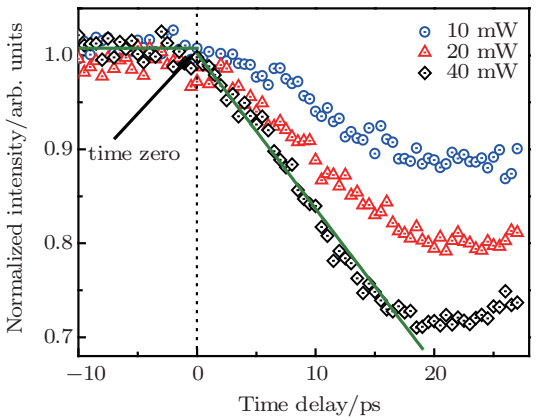


Fig. 3. (color online) The intensity evolutions of ultrafast electron bunch transverse profiles with time delay at different excitation laser intensities.

For the pump–probe scheme, the measured time constant τ is the convolution of several factors including the durations of the pump laser pulse ($\tau_{\text{laser}} = 50$ fs) and probe electron bunches (τ_{ele}), the effective buildup time of the plasma electric field (τ_{plasma}), and the temporal mismatch (τ_{mismatch}) arising from the geometry arrangement of our experimental setup.[17,23] The magnitude of τ_{plasma} is independent of the pump laser intensity and will keep constant, and τ_{mismatch} is calculated according to the following equation:[24]

$$\tau_{\text{mismatch}} = \frac{\omega_{\text{e}} \sin\theta}{c}, \tag{1}$$

where ω_{e} is the electron beam size, θ is the crossing angle between laser and electron beams, and c is the speed of light in vacuum. The value of τ_{mismatch} is estimated to be 1.2 ps with 500-μm e-beam size and 45° crossing angle configuration.

Figure 4 shows the normalized intensity evolution of ultrafast electron bunch transverse profile with time delay, caused by the plasma electric field at different electron numbers per pulse (4×10^3, 1.3×10^4, 2.96×10^4, and 5.48×10^4 electrons per pulse respectively). The time constants τ are fitted to be 3.6, 7.1, 11.0, and 14.7 ps at the electron numbers of 4×10^3, 1.3×10^4, 2.96×10^4, and 5.48×10^4 respectively. Obviously, the time constant τ shows a strong dependence on electron number as shown in the inset of Fig. 4. Moreover, their electron pulse-widths are measured to be 1.3, 2.4, 3.84, and 5.33 ps, respectively, by the autocorrelation method. Figure 5 shows the dependence of the time constant τ on ultrafast electron pulse-width. At low electron number, the time constant τ is in a similar temporal scale to τ_{mismatch}, τ_{plasma}, and τ_{ele}. With electron number increasing, the pulse-width is largely broadened due to the SC effect, and τ_{ele} becomes several times larger than τ_{mismatch} and τ_{plasma}. The experimental results shows that the time constant τ is measured to be 24.6 ps at the sample position with 10^5 electrons per pulse, corresponding to the electron pulse-width of 20.7 ps.

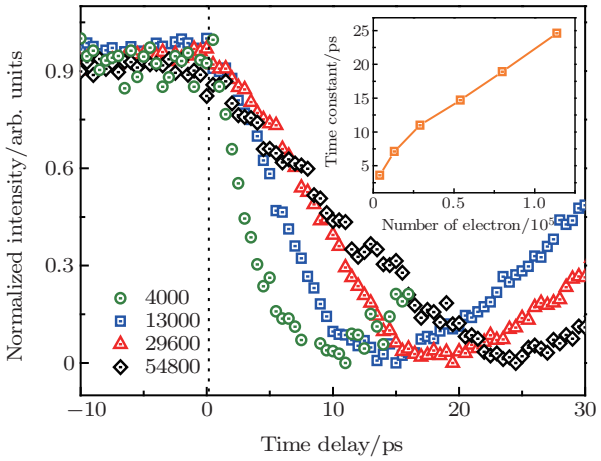


Fig. 4. (color online) The normalized intensity evolutions of ultrafast electron bunch transverse profiles with time delay at different electron numbers. The inset shows the dependence of the time constant τ on the electron number per pulse.

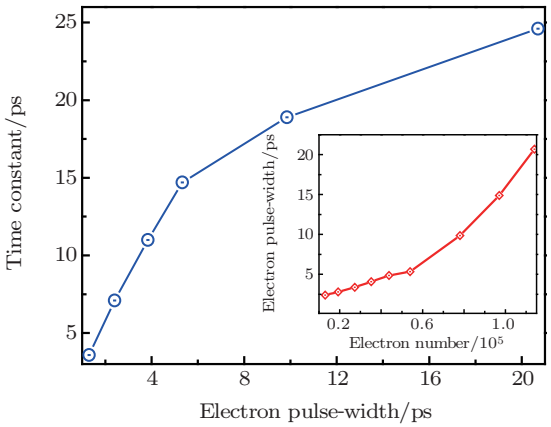


Fig. 5. (color online) Time constant τ as a function of ultrafast electron pulse-width. The inset shows the dependence of the electron pulse-width on electron number per pulse.

To obtain a single diffraction pattern with high signal-to-noise ratio, the electron number per pulse should reach a level of 10^5. Simulation shows that the SC effect results in a linear velocity chirp of ultrafast electron bunches during propagation, which make it possible to be compressed by a phase-locked sinusoidal RF electric field.[16] In our experiment, a 3.2-GHz RF source, which is phase-locked to the femtosecond laser oscillator system (Tsunami, Spectra-Physics), is introduced into the cylindrically symmetric RF cavity working

at TM_{010} mode. The phase-locked RF electric field can retard the front electrons and accelerate the back ones, and so the linear velocity chirp of ultrafast electron bunches will be pre-compensated for. At the same time, there is a transverse defocusing effect at the RF cavity entrance and exit, which can be compensated for by the magnetic lens. The momentum difference obtained by the electrons can be expressed as[25]

$$\Delta p \approx \frac{e\omega\tau_e d}{v_e} E_0 \cos\varphi_0, \tag{2}$$

where e is the elementary charge, ω is the angular frequency of the RF field, τ_e is the pulse-width of ultrafast electron bunches, d is the effective length of the RF cavity, E_0 and ϕ_0 are the amplitude and phase of the RF electric field, and v_e is the central electron velocity of ultrafast electron bunches. Based on Eq. (2), it can be estimated that ultrafast electron bunches with an energy of 50 keV and the pulse-width of 20 ps can be compressed for by a 3.2-GHz RF field with an amplitude of 0.8 MV/m. According to the particle tracing simulation (GPT),[26] ultrafast electron bunches with the electron number of up to 10^5 and a pulse-width of 20 ps can be compressed to 150 fs.

Figure 6 shows the temporal evolutions of the normalized intensity of the electron transverse profiles with and without RF compression. The measured electrons per pulse is 10^5, and the time constants are obtained to be 25 ps by fitting for no RF compression and 5.6 ps for the optimal compression phase and amplitude, respectively. It can be inferred that ultrafast electron bunches have been compressed for from 20 ps to less than 1.8 ps, down to the temporal resolution measuring limit. According to the simulation, ultrafast electron bunches can be compressed to a hundred-femtosecond time scale. Note that, the further improvement in laser and electron beam size as well as their crossing angle will be possible to measure the electron pulse-width with sub-picosecond precision.

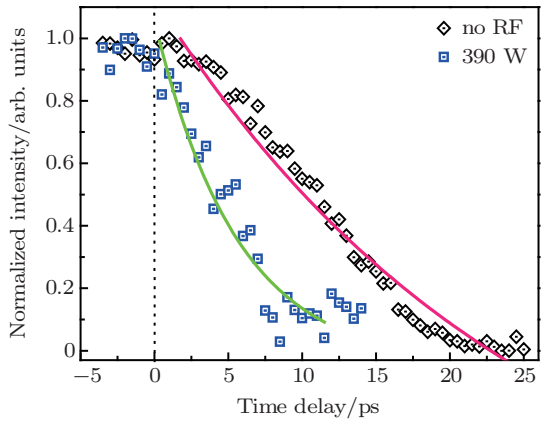


Fig. 6. (color online) Normalized intensity evolutions of ultrafast electron bunch transverse profiles with time delay with and without RF compression.

5. Conclusion

In this paper, we demonstrate a simple method of characterizing the high-brightness RF compressed electron pulse by the cross-correlation technique involving electrons and laser-induced plasma. The evolutions of electron bunch profiles repelled by laser-induced plasma indicate that the intensity decay time is strongly dependent on the electron pulse-width. The decay time decreases from 25 ps to 5.6 ps after RF compression, that is, the electron pulse-width is compressed from 20 ps to less than 1.8 ps, down to the temporal resolution measuring limit. It can be further improved to measure ultrafast electron bunches with sub-picosecond pulse-width. The experimental results indicate that the high-brightness RF compressed ultrafast electron bunches can be characterized with the temporal resolution of picosecond or down to sub-picosecond in a keV energy range, which is an optimal source for single-short electron diffraction.

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