Compressing ultrafast electron pulse by radio frequency cavity*

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An ultrafast electron diffraction technique with both high temporal and spatial resolution has been shown to be a powerful tool to observe the material transient structural change on an atomic scale. The space charge forces in a multi-electron bunch will greatly broaden the electron pulse width, and therefore limit the temporal resolution of the high brightness electron pulse. Here in this work, we design an ultrafast electron diffraction system, and utilize a radio frequency cavity to realize the ultrafast electron pulse compression. We experimentally demonstrate that the stretched electron pulse width of 14.98 ps with an electron energy of 40 keV and the electron number of 1.0×10^5 can be maximally compressed to about 0.61 ps for single-pulse measurement and 2.48 ps for multi-pulse measurement by using a 3.2-GHz radiofrequency cavity. We also theoretically and experimentally analyze the parameters influencing the electron pulse compression efficiency for single- and multi-pulse measurements by considering radiofrequency field time jitter, electron pulse time jitter and their relative time jitter. We suggest that increasing the electron energy or shortening the distance between the compression cavity and the streak cavity can further improve the electron pulse compression efficiency. These experimental and theoretical results are very helpful for designing the ultrafast electron diffraction experiment equipment and compressing the ultrafast electron pulse width in a future study.

Keywords: ultrafast electron diffraction, electron pulse compression, radio frequency cavity

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1. Introduction

As is well known, the direct observation of the atomic motion in a structural transition is a great challenge in science. In order to have a complete description of this interesting natural phenomenon, one should obtain the relevant information about both the structure and dynamics on temporal and spatial scales. Usually, the ultrafast phenomenon occurs on a temporal scale as fast as several picoseconds and even tens or hundreds of femtoseconds, while the spatial scale of some microstructures is as small as a nanometer and even an angstrom meter, which means that there is a high requirement for the relevant research methods in the observation of the ultrafast dynamics process. With the advent of the ultrafast laser technology, the observation of the ultrafast process on a microscopic scale is realized. The pump-probe technique based on the ultrafast laser pulse has been used to study the electronic dynamics happening on this ultrafast temporal scale.[1] In these experiments, a femtosecond laser pulsewas split into two components, where the intense one is used to pump while the weak one is used to probe. The pump pulse is used to excite the target sample, and the time-delayed probe pulse is used to measure the laser-induced dynamics at each moment. The temporal resolutions in these experiments are usually limited by the ultrafast laser pulse width, which can be as short as less than 10 fs. Although this method can satisfy the temporal resolution requirement, this optical technique cannot provide a direct observation of the microstructure motion, for which the basic reason is that the experimental result is obtained by the optical parameter measurement.[2,3] Recently, the ultrafast electron diffraction (UED) technique, which is equipped with both the high temporal resolution of femtosecond laser pulse and the high spatial resolution of electron diffraction, can provide a very effective approach to observing the microstructure dynamics in molecules and materials at an atomic level.[4,5] With the continual development of the UED technique, it has been used to experimentally observe various ultrafast phenomena, such as lattice heating,[6–8] chemical reactions,[9] and molecular motion in organics.[10] Furthermore, this technique has been used as a powerful tool to study ultrafast dynamics in the fields of chemistry, biology, and material science.[11–15] In these studies, the ultrafast (i.e., femtosecond) laser pulse is used to trigger the atomic motion in the matter, and the ultrashort electron pulse is used to detect the microstructure motion, whose temporal scale usually ranges from tens to hundreds of femtoseconds.

The ultrafast electron pulse in the UED experiment is usu-

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ally generated by the femtosecond photoemission from a flat metal photocathode and accelerated by the high-voltage electric field. The broadband femtosecond laser spectrum and the cathode material property will produce such an initial electron pulse with an initial emission bandwidth of ~ 0.1 eV,[16,17] which results in the rapid stretching of the electron pulse in a very short propagation distance due to the initial electron velocity distribution and space charge effect, where the leading electrons are accelerated while the tailing electrons are decelerated. The stretched electron pulse will reduce the temporal resolution of the UED system, which will make it hard to directly observe the atomic motion. The space charge effect is the biggest obstacle in achieving the ultrashort electron pulse. So far, several schemes have been proposed to directly obtain the ultrashort electron pulse or recompress the stretched electron pulse in the subsequent propagation process. A special case is the single electron pulse, where there is no space charge effect since only approximately one electron exists, thus the single electron pulse width is related to the initial emission energy bandwidth, and therefore is almost the same as the femtosecond laser pulse width.[18,19] However, in the case of the multiple electron pulse, the ultrashort electron pulse width can also be directly achieved by a compact electron gun device,[20–22] which is mainly realized by shortening the electron propagation distance from the anode to the sample position and reducing the electron density. Moreover, the space charge effect can be greatly mitigated in the MeV electron bunch, and thus the high energy electron pulse width can be reduced to sub-100 fs.[23,24] In addition, a new method based on a femtosecond laser pulse shaping technique has been theoretically proposed to realize the electron pulse self-compression in a compact electron gun device.[25] One advantage of this scheme is that the electron pulse width can be shorter than the excited laser pulse width. In this work, we build a UED system and realize the electron pulse compression based on a radio frequency (RF) technique proposed by Van Oudheusden *et al.*[26] In this scheme, the electron pulse is compressed by a time-dependent on-axis electric field in an RF cavity,[27] which imposes a linear velocity chirp that is opposite to the stretched effect of the electron pulse. Therefore, the advantage of the RF compressed UED system is that the high-brightness electron bunch with sub-picosecond pulse width can be obtained, which is highly desired in the relevant applications of the UED technique. In our experiment, the stretched electron pulse width of 14.98 ps (full width at half maximum (FWHM) value) with an electron energy of 40 keV and the electron number of 1.0×10^5 can be maximally compressed to 0.61 ps for single-pulse measurement and 2.48 ps for multi-pulse measurement by using a 3.2-GHz RF cavity, and the time jitters, including the RF field time jitter, electron pulse time jitter and their relative time jitter, are considered as the decisive factor for the electron pulse compression efficiency in the RF compressed UED system by the multi-pulse measurement. Moreover, increasing the electron energy or shortening the distance between the compression cavity and the streak cavity are proposed to further improve the electron pulse compression efficiency.

2. Experimental arrangement

In our experiment, the whole RF compressed UED system is shown in Fig. 1, which consists of two components: one is a femtosecond laser system with synchronization regime (see Fig. 1(a)), and the other one is an electron gun with a direct current (DC) photoelectron accelerator, a 3.2-GHz compression cavity and a 6.4-GHz streak cavity (see Fig. 1(b)). In Fig. 1(a), the repetition rate of the Ti:sapphire laser oscillator (Tsunami, Spectra-physics) is locked to an 80-MHz RF signal generator (N5181B, Agilent) by a phase lock loop (PLL) in the laser system. The PLL device is used to change the oscillator cavity length by controlling a piezo-electric transducer (PZT), and finally realizes the repetition rate control of the laser oscillator. Moreover, the filtered 80-MHz RF signal is multiplied in frequency and amplified in power, and thus forming 400-MHz and 800-MHz amplified RF signals, then the two RF signals are separately sent to the 3.2-GHz and 6.4-GHz frequency synchronizers with the adjustable phase and power, and finally the 3.2-GHz and 6.4-GHz digital frequency synthesized pulse signals are sent into the compression cavity and streak cavity, respectively. The Tsunami laser oscillator locked to the 80-MHz signal generator is used as a seed source of an 800-nm femtosecond laser regeneration amplifier (Spitfire Ace, Spectra-physics). Here, the repetition rates of RF signal and output femtosecond laser pulse are both 1 kHz. In Fig. 1(b), the ultrafast electron pulse is generated through photoemission from a photocathode material of 20-nm Au film by using the third harmonic (266 nm, 90 fs) of the output femtosecond laser (800 nm, 50 fs). The transverse laser profile is of a Gaussian shape with a 200-μm width in beam waist (FWHM). The generated electron pulse is then accelerated to 40 keV from the photocathode to the anode by a static electric field of 4 MV/m. After the electron beam passes through the anode, a magnetic lens is applied to control the electron beam size, and a 3.2-GHz compression cavity in the TM_{010} model is used to compress the electron pulse width, which imposes a linear velocity chirp that is opposite to the stretched effect of the electron pulse, and two pairs of deflection plates are used to adjust the electron beam position on the sample. The electron pulse width is measured by a 6.4-GHz streak cavity in the TM_{110} model, and the electron number per pulse is measured by a Faraday cup.

In order to detect a single-pulse electron streak image, we use a microchannel plate (MCP) with a phosphor screen (PS)

to enhance the electron image signal, and the image can be directly captured by an electron multiplying charge coupled device (EMCCD) camera. When the electron pulse propagates in the 3.2-GHz compression cavity, the leading electrons are decelerated and the tailing electrons are accelerated. By properly controlling the electric field intensity and phase in the compression cavity, the electron pulse width can be compressed to a minimum value at the sample position. In order to avoid that the short streak image is caused by the electron velocity change, it is necessary to ensure that the electron arrival time at the streak cavity is constant, that is to say, the center position on the MCP device for the compressed electron streak image is the same as that for the uncompressed one. Here, we first determine the streak image position by adjusting the RF phase, and then find a maximal compression by varying the RF power. Actually, the electron streak image distribution is a convolution of the transverse and longitudinal electron bunch profile,[28] and so the measured electron pulse width (FWHM) on the CCD screen is given by $\tau_{\rm scr} = (\tau_x^2 + \tau_{\rm ele}^2)^{0.5}$, where τ_x is the inherent electron pulse width due to the electron transverse bunch size on the CCD screen and $\tau_{\rm ele}$ is the real electron pulse width. It is very difficult to distinguish between the measured electron pulse width $\tau_{\rm scr}$ and inherent electron pulse width τ_x when the real electron pulse width $\tau_{\rm ele}$ is very small, and therefore we use a pinhole with a diameter of 100-μm located in the front of the streak cavity to reduce the electron bunch size (i.e., τ_x) in order to improve the measurement resolution of the electron pulse width. Further reducing the diameter of the pinhole can improve the temporal resolution of pulse width measurement, but a too small pinhole will reduce the electron number through the pinhole, and even make it difficult to form a consecutive streak image.

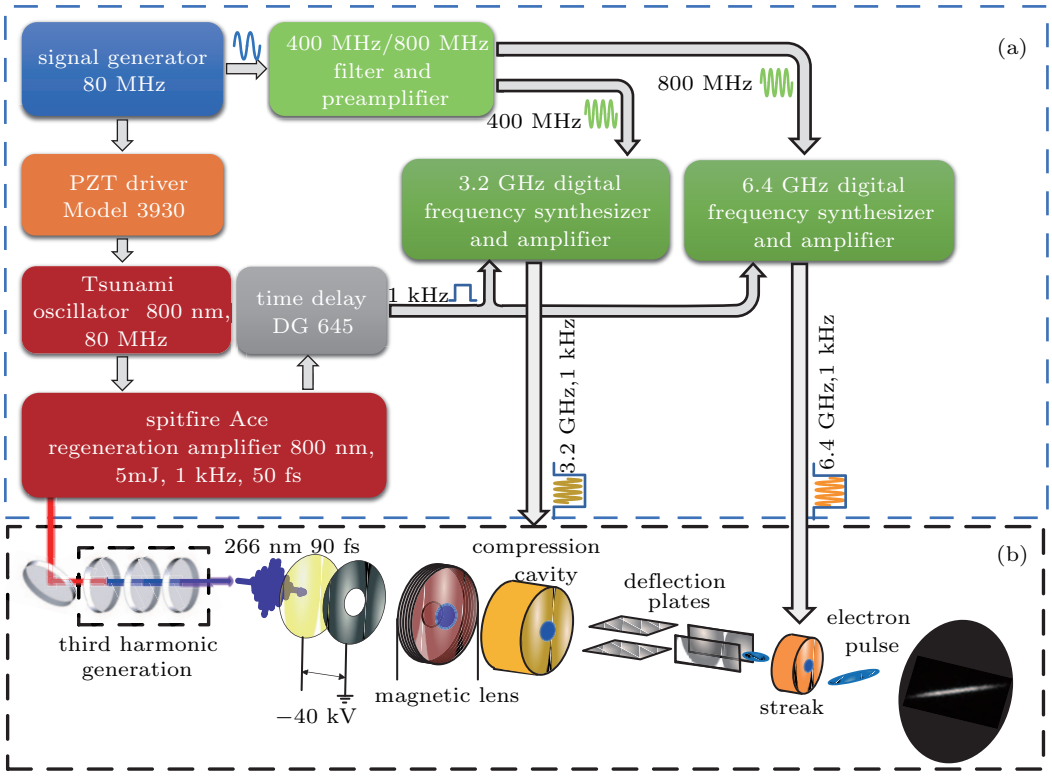


Fig. 1. (color online) Experimental arrangement of the ultrafast electron diffraction system based on a radiofrequency compression method. Panel (a) shows the block diagram of the synchronization regime, where the 80-MHz signal generator is used to generate the 3.2-GHz and 6.4-GHz frequency synchronizers by a frequency multiplication and power amplification, and is also used to lock the femtosecond laser system by a phase lock loop; panel (b) displays the schematic diagram of the electron gun, where the electron pulse is generated by back illuminating a 20-nm thick gold film, accelerated to 40 keV by a static electric field, focused by a magnetic lens, and finally compressed by a compression cavity. The electron pulse width is measured by a streak cavity combining with CCD.

3. Results and discussion

In our experiment, both the 3.2-GHz compression and 6.4-GHz streak cavities are employed with using the RF field. The 3.2-GHz compression cavity provides a time varying electric field $E_z(z,t) = E_0(z)\cos(2\pi ft)$ in the longitudinal direction, while the 6.4-GHz streak cavity provides a time varying magnetic field $B_y(z,t) = B_0(z)\cos(2\pi ft)$ in the x direction. Usually, the electric or magnetic field intensity is difficult to directly measure within the experimental setup, and so we use an RF power meter (N1912A, Agilent) to measure the RF field intensity before the RF signal is sent into the two cavities. The maximal RF powers of the 3.2-GHz compression cavity and the 6.4-GHz streak cavity are 1500 W and 1000 W in our experimental system, respectively. The temporal resolution of

the streak cavity is correlated with the RF power, the electron velocity and the distance between the streak cavity and the MCP device, and is determined to be 113 fs in our experimental condition.

In order to demonstrate the ultrafast electron pulse compression by the RF compression cavity, we measure the electron pulse width for non-compression and maximal compression cases, and the experimental results are shown in Fig. 2, where the electron number is 1×10^5. Considering the electron bunch jitter on the MCP device, we measure the electron pulse width by both the single- and multi-pulse measurements. In this experiment, the multi-pulse measurement is the integration of ten-pulse measurements. The effect of the electron bunch jitter on the non-compressed electron pulse width can be neglected due to the very wide electron pulse, and so here only the non-compressed electron pulse width obtained by the multi-pulse measurement is given (see Fig. 2(a)). In the case of non-compression, as shown in Figs. 2(a) and 2(e), the measured electron pulse width is stretched to about $\tau_{\rm scr} = 15.01$ ps due to the space charge effect. However, in the case of maximal compression, as shown in Figs. 2(c), 2(d), and 2(f), the measured electron pulse widths are compressed to $\tau_{\rm scr} = 1.07$ ps and 2.61 ps for single- and multi-pulse measurements, respectively. The maximum compression of the electron pulse width is achieved in an RF power of 35 W, but our experimental system can provide a maximal RF power of 1500 W, and so the electron pulse with the higher electron energy and a bigger electron number can also be compressed in our experimental system. Additionally, as shown in Figs. 2(b) and 2(f), the inherent electron pulse width due to the electron bunch size is $\tau_x = 0.88$ ps. Based on such a relationship $\tau_{\rm ele} = (\tau_{\rm scr}^2 - \tau_x^2)^{0.5}$, the real non-compressed electron pulse width can be calculated to be 14.98 ps, while the real maximally-compressed electron pulse widths are 0.61 ps and 2.48 ps for single- and multi-pulse measurements, respectively. One can see that the electron pulse width can be compressed by a factor of ~ 25 for the single-pulse measurement. Moreover, the real electron pulse width for the single-pulse measurement is much smaller than that for the multi-pulse measurement, and the fundamental reason is that the multi-pulse measurement involves the electron bunch jitter on the MCP device, which results from the RF field time jitter, electron pulse time jitter and their relative time jitter.

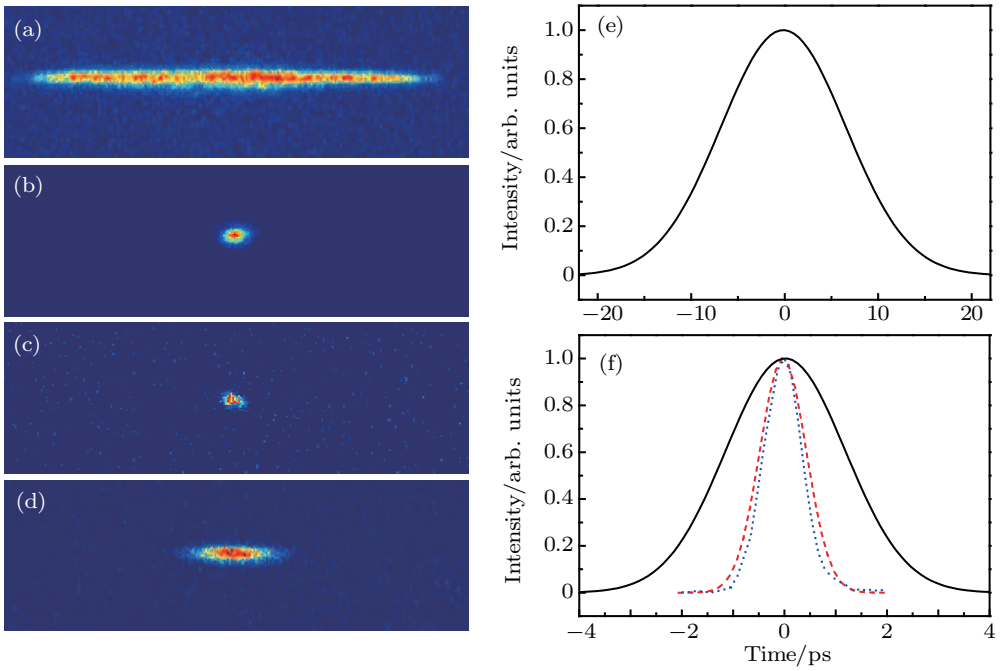


Fig. 2. (color online) Streak images for non-compressed electron bunch with (a) and without (b) the streak cavity and maximally compressed electron bunch with single- (c) and multi-pulse (d) measurements, together with the calculated electron pulse profiles for non-compressed electron bunch with (e) and without (blue dotted line) (f) the streak cavity, and the maximally compressed electron bunch with single- (red dash line) and multi-pulse (black solid line) measurements (f).

In order to show the maximal compression efficiency of the ultrafast electron pulse for the single-pulse measurement in our experimental system, we theoretically simulate the electron pulse width and electron beam size evolution behavior in the propagation process according to our experimental condition, and the calculated results are shown in Fig. 3, where the space charge effect has been taken into account in the whole electron pulse trajectory. As can be seen, both the electron beam size and electron pulse width are rapidly increased after the photocathode ($z = 0$ m), then the electron beam size is reduced after the magnetic lens ($z = 0.17$ m), while the electron pulse width is reduced after the compression cavity ($z = 0.29$ m), and finally both the electron beam size and electron pulse width are maximally reduced as much as possible

at the sample position. In our case, the maximally compressed electron pulse width is 0.57 ps, and the electron beam size is 400 μm in diameter. Obviously, our experimental value of 0.61 ps is close to the theoretical calculation, which indicates that our experimental system has basically reached the optimal condition. The longitudinal pulse width and the transverse beam size are two important factors in the spatiotemporal resolution of the UED system, but it is difficult to simultaneously make the electron pulse width and electron beam size achieve their minimum values, and the main reason is that the compression cavity will also affect the electron beam size. An alternative method is to design two magnetic lenses, one is to collimate the electron beam in the front of the compression cavity, and the other is to focus the electron beam in the back of the compression cavity, thus we can obtain both the short electron pulse width and small electron beam size.

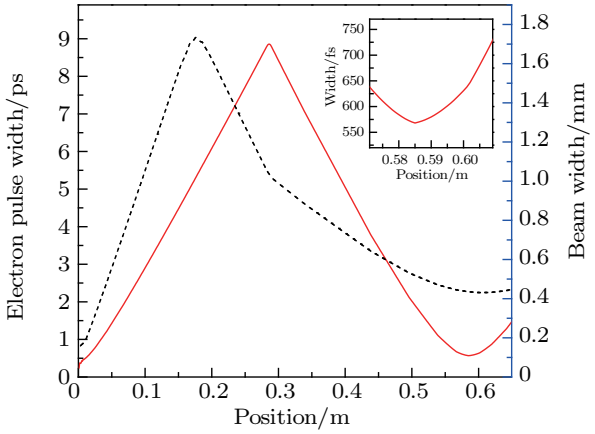


Fig. 3. (color online) Theoretical simulations of electron pulse width (red solid line) and beam diameter (black dash line) as a function of the propagation distance z from the cathode. The inset shows the minimal electron pulse width around the sample position $z = 0.59$ m.

In several experiments using the UED technique, the multi-pulse measurement is necessary in order to obtain the clear electron diffraction images. Usually, the temporal resolutions in the UED experiments will be strongly influenced by the arrival times of these electron pulses. The 3.2-GHz RF field in the compression cavity is an important generation source for the electron pulse time jitter in the multi-pulse measurement. The phase jitter of the RF signal will cause the electron bunch velocity to change, and finally result in the arrival time jitter at the sample position. Next we measure the phase noise (i.e., time jitter) of 3.2-GHz RF field in the compression cavity by a signal source analyzer (E5052B, Agilent), and the measurement result is shown in Fig. 4. As can be seen, the time jitter of the 3.2-GHz RF field is $\sigma_{\mathrm{RF}} = (0.3 \pm 0.01)$ ps, which almost keeps the same value of the 80-MHz RF signal generator (N5181B, Agilent) in a frequency range of 1 Hz–1 MHz. The time jitter of electron pulses due to the femtosecond laser pulse jitter is $\sigma_{\mathrm{ele}} = 0.4$ ps. Thus, the relative time jitter between the electron pulse and the 3.2-GHz RF field can be calculated as follows: $\sigma_{\mathrm{rel}} = (\sigma_{\mathrm{RF}}^2 + \sigma_{\mathrm{ele}}^2)^{0.5} = 0.5$ ps. Considering the Gaussian profile, the relative time jitter (FWHM) is $2.355 \times \sigma_{\mathrm{rel}} \approx 1.17$ ps.

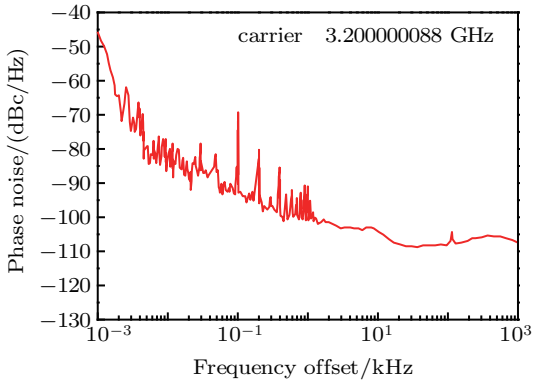


Fig. 4. (color online) Phase noise (i.e., time jitter) of the 3.2-GHz RF field in the compression cavity, measured by a signal source analyzer (E5052B, Agilent).

The relative time jitter between the electron pulse and the 3.2-GHz RF field is a critical effect parameter for the ultrafast electron pulse width in the multi-pulse measurement. We theoretically analyze the effects of the relative time jitter on electron pulse width jitter and arrival time jitter. Figure 5 shows the simulation results of the pulse width jitter and arrival time jitter at the sample position as a function of the relative time jitter. One can see that the relative time jitter of 1 ps will lead to a pulse width jitter of 2.5 fs and an arrival time jitter of 2.2 ps. Obviously, the relative time jitter almost does not affect the electron pulse width, but will greatly influence the electron arrival time. Basically, several-femtosecond pulse width jitter can be ignored in those sub-picosecond UED experiments. However, the arrival time jitter will be greatly amplified, and the amplification coefficient is about 2.2 based on our linear fitting. In our experiment, the relative time jitter is 1.17 ps, and thus it will lead to an arrival time jitter of about 2.57 ps at the sample position. An alternative method to reduce this amplification coefficient is to shorten the distance between the compression cavity and the streak cavity (i.e., sample position).

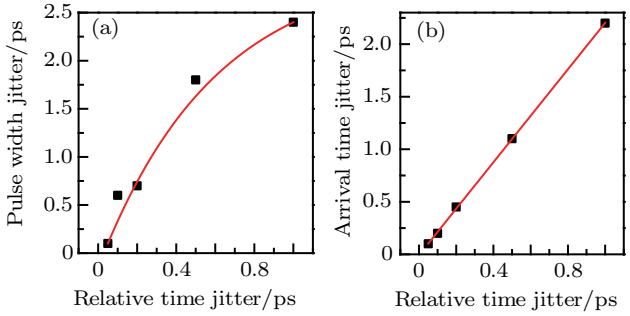


Fig. 5. (color online) (a) Electron pulse width jitter and (b) arrival time jitter at the sample position as a function of the relative time jitter (black squares), together with the theoretical fitting (red solid lines).

Finally, we theoretically and experimentally study the maximally compressed electron pulse width by varying the RF

power for the multi-pulse measurement. Figure 6 shows the measured electron pulse width as a function of the 3.2-GHz RF field power with the 10-pulse measurement, together with the theoretical simulation. It can be seen that the experimental data can be in good agreement with the theoretical simulations, and the electron pulse width can be maximally compressed to 2.61 ps (measured value) with the RF field power of 35 W as shown in Fig. 2. Furthermore, we also measure the electron pulse width with the electron number of 3000, and the maximally compressed electron pulse width is also about 2.61 ps, which indicates that the arrival time jitter has a great influence on the electron pulse width for the multi-pulse measurement. In order to further improve the compression efficiency of electron pulse width, next, we plan to make some improvements in our UED setup, for example, we can increase the electron energy or shorten the distance between the compression cavity and streak cavity, and thus these electrons have the higher propagation speed and shorter propagation time. Therefore, such an electron bunch with a pulse width of sub-100 fs is expected to be obtained.

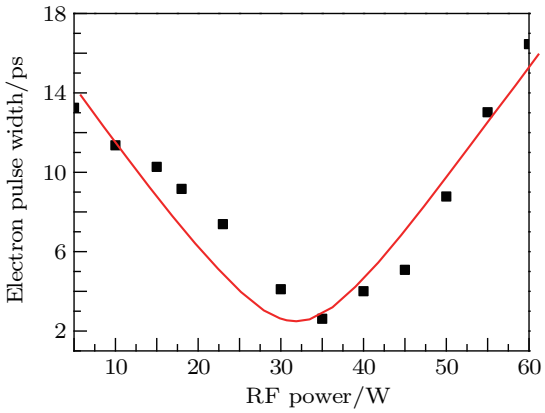


Fig. 6. (color online) Electron pulse width as a function of the RF power of the compression cavity with the 10-pulse measurement (black squares), together with the theoretical simulation (red solid line).

4. Conclusions

In this work, we demonstrate that the RF cavity can provide an effective method to recompress the ultrafast electron pulse in the UED system. Our experimental results show that the stretched electron pulse width of 14.98 ps with the electron energy of 40 keV and electron number of 1.0×10^5 can be maximally compressed to 0.61 ps with single-pulse measurement, and 2.48 ps with multi-pulse measurement. Our theoretical and experimental analyses indicate that the RF field time jitter, electron pulse time jitter and their relative time jitter are the main parameters to affect the electron pulse compression efficiency for the multi-pulse measurement. Additionally, increasing the electron energy and shortening the distance between the compression cavity and the streak cavity are shown to be two feasible schemes to further improve the electron pulse compression efficiency. These studies present a clear physical picture for the electron pulse broadening and compression in the propagation process, which can provide the experimental and theoretical basis for properly designing the UED system to obtain the ultrashort electron pulse.

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