Coulomb interaction-induced jitter amplification in RF-compressed high-brightness electron source ultrafast electron diffraction

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Abstract

We have theoretically and experimentally demonstrated an RF compression-based jitter-amplification effect in high-brightness electron source ultrafast electron diffraction (UED), which degrades the temporal resolution significantly. A detailed analysis and simulations reveal the crucial role of the longitudinal and transverse Coulomb interaction for this jitter-amplification effect, which accord very well with experimental results. An optimized compact UED structure for full compression has been proposed, which can suppress the jitter by half and improve the temporal resolution to sub-100 fs. This Coulomb interaction-induced jitter amplification exists in nearly the whole ultrafast physics field where laser–electron synchronization is required. Moreover, it cannot be suppressed completely. The quantified explanation for the mechanism and optimization provides important guidance for photocathode accelerators and other compression-based ultrashort electron pulse generation and precise control.

1. Introduction

Utilizing ultrafast optical and free-electron pulse systems together, the observation of the ultrafast changes in many scientific fields such as chemistry \cite{1–3}, material sciences \cite{4, 5}, and condensed matter physics \cite{6, 7} can be achieved in real time. In particular, this approach can determine the structures and dynamics of most intermediate phenomena with sub–picosecond temporal and sub–angstrom spatial resolution and describe the entire process in unprecedented detail. Ultrafast electron diffraction (UED) is one of the promising techniques based on a pump and probe scheme. In UED, an ultrashort laser pulse is used to excite the sample and then at various time delays, the corresponding dynamics is probed by an ultrashort electron bunch \cite{8–13}.

Recently, ‘high-brightness’ UED \cite{14}, i.e. the UED system with a high-brightness electron source, has provided a pathway towards the imaging of irreversible and cumulative heat-effected ultrafast dynamics. Here, the brightness is defined in terms of the electron bunch density and the transverse coherence \cite{14}. The greater the number of electrons in a pulse and the shorter the electron pulse duration, the brighter the electron source of the UED. However, the Coulomb repulsion between electrons will broaden the electron pulse in the transverse and longitudinal directions, and greatly expand the electron pulse duration. An ultrafast time-dependent electric field is needed to compensate the expansion for generating ultrashort multi-electron pulses. RF technique has been widely used in the fields of accelerator physics and electron pulse control (e.g. photocathode as the electron source), and has also been introduced to UED for both single-electron pulse compression mode \cite{15} and high-brightness compression mode \cite{16}. In general, the temporal resolution of the pump-probe technique in RF-compressed high-brightness electron source UED depends on the laser pulse duration, the electron pulse duration, and their relative timing jitter \cite{17–19}. Nowadays, sub–10 fs FWHM laser pulse from the ultraviolet to
near-infrared region can be directly obtained from a commercial Ti:sapphire laser system, and the extreme ultraviolet attosecond pulse can also be experimentally realized by high harmonic generation [20]. Therefore, the temporal resolution is just limited by the electron pulse duration and the relative timing jitter. Recently, it has been demonstrated that an RF cavity with TM010 mode can be used to reverse the Coulomb interaction-induced linear energy chirp [21] and compress the electron pulse down to sub-100 fs FWHM duration [22, 23]. This RF compression is based on the ballistic compression regime when the electron pulse passes the center of the cavity, and the RF field in the compression cavity is tuned just at the time of zero voltage. After compression, the average energy of the electron pulse will not change. Compared with the ballistic compression, the RF velocity bunching scheme accelerates or decelerates the electron pulse after compression, which provides the generation of the shortest possible high-brightness electron pulse. This velocity bunching scheme is also widely investigated in many fields, including KeV UED [24], MeV UED [25], and ultrafast electron microscopy (UEM) [26].

Although the RF compression can provide the shortest electron pulse, the timing jitter problem in RF compression limits the temporal resolution, which cannot be avoided. For RF-compressed high-brightness electron source UED, in order to inject an electron pulse into a compression cavity on the desired phase of the RF field, the RF field should be synchronized to the femtosecond laser pulse, which generates the photoelectrons (i.e. the electron pulse). Laser-microwave synchronizations with a few femtosecond jitter, or even hundreds of attosecond jitter have been achieved where the laser works at the repetition rate of a few to tens of MHz [27–29]. However, in this case, the laser pulse energy is insufficient to excite the structural dynamics in UED experiments. Therefore, synchronization of microwaves to a laser amplifier system (normally, kHz repetition rate, ~10 μJ pulse energy) is required and widely explored in UED experiments. The huge time-scale difference between the microwave period and laser repetition rate makes it difficult to achieve sub-100 fs rms synchronization jitter. Based on the phase-locked loop (PLL) system, a 3 GHz electronic oscillator is successfully synchronized to a mode-locked Ti:sapphire laser (a 75 MHz seed laser, which feeds into a kHz regenerative amplifier) with less than 20 fs rms phase jitter (0.05 Hz–100 KHz bandwidth) [30]. For this synchronization scheme, a direct measurement result of the synchronization jitter between the femtosecond laser pulse and electric field inside the RF cavity is ~100 fs (rms) [31]. This synchronization jitter at the cavity position is also known as the relative timing jitter. With the same synchronization regime, the measured jitter is ~200 fs rms [32] at the sample position (i.e. the temporal focus). This synchronization jitter at the sample position is known as the arrival timing jitter. Although the difference in jitter measurement results between the relative timing jitter at the cavity position in [31] and the arrival timing jitter at the sample position in [32] is large, the mechanism is ambiguous. The similar different jitter measurement results between the relative timing jitter and arrival timing jitter can also be found in the synchronization system in MeV UED. Although the relative timing jitter between the laser and electric field has been well investigated, the system structure influence for the jitter performance, i.e. the arrival timing jitter, is overlooked and has never been explored quantitatively.

In this paper, we demonstrate the Coulomb interaction-induced jitter-amplification effect in a high-brightness electron source UED system through experiment and theoretical simulation. The quantitative analysis shows that the Coulomb force significantly enlarges the synchronization jitter, i.e. the arrival timing jitter at the sample position is larger than the relative timing jitter at the cavity. This so-called jitter-amplification effect provides an unambiguous interpretation, which comprehensively explains the existing confusion regarding different jitter measurement results reported in the literature. The proposed optimization methods and criterion for the definition of this jitter-amplification elimination make the RF technique more robust and precise.

2. High-brightness electron source UED system and synchronization scheme

We have home-built a compression cavity and a deflection cavity for electron pulse compression and pulse duration measurement. The specifications of the above-mentioned cavities are tested with a network analyzer and summarized in table 1. The synchronization system in our setup is shown in figure 1(a). The Ti: sapphire oscillator is locked to a signal generator (Agilent N5181B) at 80 MHz by a PLL in the laser system. As the repetition rate of the laser is inversely proportional to the cavity length, the PLL servo controls the piezo-electric transducer to tune the cavity length for compensating the jitter and drift of the laser oscillator. The jitter of the signal generator is 700 ± 10 fs rms in the frequency range of 1 Hz–10 MHz at 80 MHz center frequency, which is much better than the jitter and drift of a laser oscillator. As for the RF signal generation, a narrow band filter centered at 400 MHz is used to select the 5th harmonic of the signal source, and then this harmonic signal is amplified and sent to the digital frequency synchronizer as a reference clock. The 3.2 and 6.4 GHz signals from the digital frequency synthesizer, which have nearly the same absolute jitter as that of the signal generator, are fed into the RF compression and deflection cavity, respectively.
The RF compression-based high-brightness electron source UED setup is shown in figure 1(c). The 90 fs (FWHM), 800 nm, 1 kHz laser pulse passes through the frequency tripling system and generates a 266 nm laser pulse. Then, the 266 nm laser pulse irradiates the 20 nm thin gold photocathode and generates the corresponding photoelectron pulse. Several fC electrons per pulse with 190 μm diameter (FWHM) are accelerated to 40 keV through an 8 mm gap. The electron pulse is focused and collimated into the RF compression cavity by a magnetic lens. The electron pulse energy chirp induced by Coulomb interaction during propagation is reversed in the compression cavity. As a result, the shortest pulse duration can be achieved at the sample position (i.e. the temporal focus position). We installed a streak cavity at the sample position to measure the electron pulse duration and timing jitter.

Table 1. Properties of the deflection and compression cavity with a reflection measurement.

| Parameter                        | RF deflection cavity | RF compression cavity |
|----------------------------------|----------------------|-----------------------|
| Center frequency f (MHz)         | 6402.0               | 3201.0                |
| Bandwidth BW (kHz)               | 900                  | 600                   |
| Q-factor                         | 14 222               | 10 666                |
| Cavity impedance                 | 44.5                 | 46.5                  |
| Standing-wave ratio              | 1.21                 | 1.09                  |

Figure 1. The schematic of the high-brightness electron source UED system. (a) The block diagram of the synchronization regime. The laser is locked to the 80 MHz signal generator by PLL with <0.5 ps rms timing jitter. The 3.2 and 6.4 GHz RF signals have the same level phase jitter with the 80 MHz signal generator. The RF signal and laser pulse in (b) determine the relative timing jitter. (c) The UED beamline structure. Electron pulses are produced by back-illumination of a gold cathode (z = 0 cm). Then, they are accelerated to 40 keV through an 8 mm gap. The magnetic lens (z = 17 cm) and deflection plates focus and collimate the electron pulse to pass through the compression cavity (z = 29 cm) and deflection cavity (z = 59 cm, i.e. the sample position). The arrival timing jitter is the electron pulse arrival time fluctuation at the sample position.
3. Electron pulse duration and jitter-amplification effect analysis

Our experiments are executed in an accumulation mode, and so the measured electron pulse duration involves the jitter. After decoupling, the jitter information can be separated from the measured electron pulse duration. Before measuring the electron pulse duration, we first characterize the streak velocity of the deflection cavity. Operating the streak cavity for 5 fC electrons per pulse, the entire pulse is shifted parallel to the streaking direction by varying the time delay arm and then recording the displacement of the center of the electron pulse by an EMCCD camera [32]. The accurate displacement is obtained by Gaussian fitting of each pattern and every Gaussian-fitted pattern accumulates for 5 s. As shown in figure 2, the streak velocity is $\sim 100$ fs/pixel and the error $4$ fs/pixel is mainly attributed to the laser-spot fluctuation on the photocathode, which is relatively small and can be neglected for the following measurements. As for the influence from the jitter between the deflection cavity RF field and the electron pulse, the center position of the measured streak pattern will not be altered for an integrated Gaussian distribution, and so this influence can be neglected. Before measuring the compressed electron pulse duration, we tested and calibrated the system through measuring the free-propagated-electron pulse duration, we take 60 000 electrons per pulse, which are located at the linear part of the photocathode quantum efficiency. The measurement results of the compressed electron pulse duration are shown in figure 3(a) with a streak velocity of 138 fs/pixel. The shortest duration is $\sim 2.6$ ps FWHM (FWHM = 2.355$\sigma_{\text{rms}}$ for Gaussian distribution) after a deconvolution of the influence from the transverse spot size. For the same condition, the simulation result calculated by GPT is shown in figure 3(b). The measured shortest pulse duration after compression is much larger than the simulation result $\sigma_{\text{ele}} = 0.36$ ps FWHM. The measurement result involves the electron pulse duration and the electron pulse arrival timing jitter at the sample position. For our synchronization regime, the RF phase jitter $\sigma_{\text{RF}}$ is 300 fs rms (corresponding to the 600 kHz bandwidth of the compression cavity) and the laser pulse jitter $\sigma_{\text{laser}} = 400$ fs rms. If we assume they are independent, the relative timing jitter between the laser pulse and RF phase at the compression cavity is $\sigma_{\text{rel}} = \sqrt{\sigma_{\text{RF}}^2 + \sigma_{\text{laser}}^2} = 500$ fs (rms). The electron pulse copies the laser pulse jitter for the sub-femtosecond photoelectron generation process, and so the relative timing jitter between the electron pulse and RF phase at the cavity position is 500 fs rms. The electron pulse duration and relative timing jitter coupled result

$\sqrt{\sigma_{\text{ele}}^2 + (\sigma_{\text{rel}} + 2.355)^2} = 1.22$ ps FWHM is still much smaller than the real measurement 2.6 ps FWHM at the sample position. The relative timing jitter-induced electron pulse duration change after compression is several fs under this condition and can be neglected. Although the arrival timing jitter is derived from the relative timing jitter, the arrival timing jitter at the sample position (i.e. the temporal focus) finally determines the temporal resolution in UED dynamics experiments. The electron pulse arrival timing jitter at the sample
position may not be equal to the relative timing jitter at the cavity, particularly for high-brightness electron pulse involving strong Coulomb interaction during propagation.

The relative timing jitter between the RF phase and laser pulse leads to the change of the electron pulse velocity and so influences the arrival time at the sample position. As a result, the relative timing jitter at the cavity position transfers to arrival timing jitter at the sample position. According to the analytical expression from Pasmans [36], the electron pulse arrival timing jitter at the sample position is equal to the RF phase jitter (i.e. relative timing jitter, which can be transformed to the RF phase jitter) under the condition of optimal compression ($\phi = \pi / 2$) and full compression where the space-charge force can be neglected. That is to say, the arrival timing jitter changes linearly with the phase fluctuation of the RF compression field. Full compression means that the longitudinal momentum change of the electron pulse from the RF compression field is large enough so that the Coulomb interaction-based resistance can be neglected for subsequent propagation. In this case, the multi-electron pulse compression is equal to single-electron pulse compression for which the arrival timing jitter is equal to the relative timing jitter. However, the so-called fully compressed electron pulse would not be guaranteed for the common experimental condition and cannot be realized completely. Under the moderate electron pulse compression condition, the arrival timing jitter will not be equal to the relative timing jitter. In principle, the longitudinal space-charge force and the transverse focusing will resist the compression, and then the temporal focus position will be shifted backwards and deviate from the theoretical condition. In this case, the arrival timing jitter at the sample position is not equal to but larger than the relative timing jitter, which can be considered as a jitter-amplification effect.

We then simulate this amplification effect for our UED experimental setup with GPT. The electric fields in the dc accelerator, RF compression cavity, and the magnetic field in the magnetic lens are calculated with Poisson Superfish. The simulated evolution of the electron pulse is shown in figure 3(b) and the corresponding linear relationship between the relative timing jitter and arrival timing jitter for 0.54 A magnetic lens current is shown in figure 3(c). In this case, the RF field phase fluctuation (i.e. the relative timing jitter) $\Delta \phi_0 = 300$ fs at the compression cavity will induce $\sim 660$ fs arrival timing jitter at the sample position and the arrival timing jitter is

Figure 3. The compression results from experiments and simulation with GPT. (a) The electron pulse duration as a function of the RF power, and the red line is the fitting curve. For 138 fs/pixel streak velocity, the shortest pulse duration is $\sim 2.6$ ps FWHM. (b) The simulation results of the evolution of the compressed electron pulse duration (the black symbols) together with the diameter profile (the blue symbols) as a function of the propagation distance from the cathode. The circles are for 0.54 A magnetic lens current and the stars are for 0.59 A current. (c) The linear relationship between relative timing jitter and arrival timing jitter. The slope rate, i.e. the amplification factor, is 2.2. (d) Increasing the compression cavity field can weaken the influence of the Coulomb force. The amplification factor (the black circles) reduces, and the temporal focus position (the blue squares) moves towards the cavity position as the compression field increases. The photocathode position is the 0 cm position in this picture.
2.2 ps for $\Delta \varphi_0 = 1$ ps. So the amplification factor, which defines the ratio of the arrival timing jitter to the relative timing jitter, is 2.2. Considering the relative timing jitter $\sigma_{rel} = 500$ fs rms and the FWHM value is $2.355 \sigma_{rel}$ for Gaussian distribution, the measured pulse duration at the sample position should be $2.2 \times 2.355 \sigma_{rel} = 2.59$ ps FWHM, which agrees well with the experimental result. It should be noted that this 2.59 ps FWHM simulation result, which involves the compression cavity jitter and amplification effect, neglects the influence from deflection cavity jitter, but the experimental result involves. If we assume the compression cavity jitter and deflection cavity jitter are independent, the deflection cavity jitter is 2.355°0.5° = 1.18 ps FWHM. The deviation for with and without deflection cavity jitter is $\sqrt{2.59^2 + 1.18^2} - 2.59 = 0.25$ ps FWHM and if we consider the correlation between the compression cavity jitter and deflection cavity jitter, the deviation should be much smaller and negligible. When the magnetic lens current increases to 0.59 A, as shown in figure 3(b), the electron beam size is reduced and the amplification factor is 2.4. That is to say, the tight transverse focusing intensifies the Coulomb interaction, which enlarges the amplification factor. The weakened amplification effect can be achieved by increasing the compression field, as shown in figure 3(d). The stronger compression field will induce larger momentum change, then the Coulomb interaction-based compression resistance is weakened and the jitter-amplification factor will decrease. When full compression is reached, the jitter-amplification effect vanishes and the amplification factor is 1. Similar results can be attained from the contrast of the experimental measurements in [31, 32, 37]. For the same synchronization system, the relative timing jitter between the laser pulse and the RF field inside the cavity is ~100 fs rms in [31]. However, the compressed electron pulse arrival timing jitter is 200 fs rms for the experimental setup in [32, 37]. Consequently, the Coulomb interaction-induced moderate compression in [32, 37] is the cause of the jitter amplification, which agrees well with our above-mentioned analysis and simulation.

4. Analytical model for Coulomb interaction-induced jitter-amplification effect

The present work demonstrates the jitter-amplification effect and the experiments and simulation results fit well. We attribute this jitter-amplification effect to Coulomb interaction and provide the qualitative analysis from common sense. We then make a more direct and clear insight into the mechanism of the jitter amplification from the analytical model. Considering the compression cavity as a tunable temporal lens, the dependence of the temporal focus distance on the electric field strength of the cavity follows the formula [38]:

$$z_{focus} \approx \frac{2\sqrt{2/m} U_k^{3/2}}{e\omega d_{cv} E_0 \cos(\varphi_0)}$$

where $m$ is the mass of the electron, $U_k$ is the kinetic energy of the electron pulse, $e$ is the electron charge in Coulomb, $\omega$ is the angular frequency of the RF field, $d_{cv}$ is the effective cavity length, $\varphi_0$ is the RF field phase. This analytical expression neglects Coulomb interaction and the experimental condition involves the Coulomb interaction among electrons. The temporal focus position for our experimental condition and analytical expression are presented in figure 4. Taking the amplitude of the field $E_0$ and phase $\varphi_0$ into the formula, we get the analytical temporal focus distance from the compression cavity $d_{anal} = 13$ cm as shown in figure 4(b). According to the analytical resolution in [36], for this distance, the arrival timing jitter is equal to the RF phase jitter (i.e. the relative timing jitter). However, for our experimental condition, the temporal focus position (i.e. the distance from the cavity to the sample position) is $d_{exp} = 30$ cm, which is lengthened, because of the Coulomb repulsion, as shown in figure 4(a). For a defined electron pulse velocity after compression, the arrival time at the sample position and the propagation distance $d$ from the compression cavity position follows $t \approx z$. The relative timing jitter leads to the change of the electron pulse velocity and for the distance $d = d_{anal}$, the arrival timing jitter is equal to the relative timing jitter. In this case, for the present experimental condition, $d_{exp} = 2.3 d_{anal}$, then the arrival timing jitter is 2.3 times larger than the relative timing jitter. The simulation and analytical solution match well with the experimental amplification factor of 2.2. Indeed, if we simulate the experimental condition with GPT, but remove the space-charge effect, the simulation distance from the cavity position to the temporal focus position is exactly the same as the analytical calculation. Now, the mechanism of the jitter-amplification effect has been completely revealed. The Coulomb interaction plays the crucial role in this jitter-amplification effect.

5. Optimization for UED system to suppress the jitter-amplification effect

This jitter-amplification effect is derived from the transverse and longitudinal Coulomb interaction. The Coulomb repulsion completely resists the compression, since the energy chirp is reversed in the RF compression. The method for reducing the amplification effect is to weaken the Coulomb interaction and increase the compression field up to the so-called full compression where the Coulomb interaction can be
The enhancement of the electron pulse energy and reduction of the electron pulse density assist to weaken the Coulomb interaction. However, for a high-brightness or single-shot electron source UED system, lowering the electron pulse density is inadvisable. The enhancement of the electron pulse energy will need more magnetic lens currents to focus the transverse spot, which will enlarge the Coulomb interaction. Increasing the extraction field is also effective, but larger than 10 kV mm\(^{-1}\) extraction field is really difficult as it requires tens to hundreds of kilovolt DC electron source. The increase of the RF field drastically suppresses the jitter-amplification effect, as shown in Figure 3\((d)\) and correspondingly, the temporal focus position will move towards the cavity position. So a compact structure between the cavity and sample is the most efficient and necessary optimization method and, in addition, a much shorter pulse duration can be achieved, as shown in Figure 4\((c)\).

6. The influence of compression quality

The jitter and electron pulse duration determine the temporal resolution of the UED system. Our work demonstrates the jitter-amplification effect, for which the arrival timing jitter is larger than the relative timing jitter. It should be noted that the timing jitter indeed may also influence the compressed pulse duration. For the above-mentioned simulation condition, 1 ps phase shift of the RF field will induce a several fs change of the compression duration and this change can be neglected compared with \(\sim 130\) fs rms duration at the sample position. However, if the electron pulse is compressed, sub-10 fs FWHM, the RF phase jitter-induced duration fluctuation will significantly influence the temporal resolution and should be taken into consideration.
7. Summary and discussion

We have presented the first demonstration of a jitter-amplification effect in a high-brightness electron source UED system and demonstrated that this jitter amplification is derived from the longitudinal and transverse Coulomb interaction. The dependence of the arrival timing jitter at the sample position on the relative timing jitter at the compression cavity position is linear for our system and the ratio, i.e. the amplification factor, is 2.2. A detailed analysis on the mechanism through GPT simulation and analytical expression agrees well with the experimental results. Our experimental and theoretical evidence leads to an interpretation, which comprehensively explains the existing deviations for different measurement results reported in the literature. The experiment and simulation results suggest that full compression is critical and should be guaranteed as possible not only for shorter pulse duration but to decrease the jitter-amplification factor. After the optimization provided in this work, sub-100 fs FWHM stable temporal resolution can be reached based on the synchronization and high-brightness electron source UED system mentioned above, which brings a significant promotion.

It should be noted that the energy chirp of the electron pulse before compression and the Coulomb repulsion during compression, linear or nonlinear, are all derived from Coulomb interaction. Our interpretation for the Coulomb interaction-induced jitter amplification is general and is suitable for ballistic compression and velocity bunching-based high-intensity electron beam compression schemes. Only on condition that the compression field is strong enough will the Coulomb interaction be neglected during compression, and the jitter-amplification effect will vanish. However, Coulomb interaction always exists in the multi-electron pulse, and so this Coulomb interaction-induced jitter amplification cannot be removed completely in principle. The criterion for jitter-amplification elimination is that with and without Coulomb interaction, the temporal focus position will not change or the electron pulse duration at the temporal focus will not change.

Our work on strong Coulomb interaction-induced jitter amplification provides an important guide for RF technique-based ultrashort electron pulse generation and precise control where laser–electron synchronization is needed, such as in free-electron lasers [40], the Thomson scattering x-ray source [41], MeV UED [42, 43], keV UED [32, 57], and UEM [26]. The jitter-amplification effect can be found in MeV UED [44] and before our work, indicative that the mechanism is unambiguous. The tight transverse focusing in keV and MeV UEM [26] intensifies the Coulomb interaction and the jitter-amplification factor will be enlarged, which should be taken into consideration. Although the laser [45–47] and Thz [48, 49]-based multi-electron pulse compression and diagnosis [50, 51] can neglect the synchronization jitter in principle, without appropriate design, and few femtosecond jitter times, the jitter-amplification factor will significantly hinder the way towards few femtosecond to attosecond temporal resolution. In summary, this Coulomb interaction-induced jitter amplification needs to be carefully considered for general applications requiring both a laser and multi-electron beam.

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