Femtosecond electron pulse compression by using the time focusing technique in ultrafast electron diffraction*

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We present a new model of an electron gun for generating subrelativistic femtosecond (fs) electron pulses. The basic idea is to utilize a dc acceleration stage combined with a time focusing region, the time focusing electrode generates an electron energy chirp for bunching at the target. Without considering the space charge effects, simulations of the electron gun were carried out under the conditions of different dc voltages and various slopes of the voltage added on the time focusing electrode. Tracing and simulating large numbers of photoelectrons through Monte–Carlo and finite difference methods, the electron pulses with 1 ps can be compressed to 55 fs, which will allow significant advances in the field of ultrafast diagnosis.

Keywords: time focusing technique, temporal pulse compressing, time-dependent electric field, Monte–Carlo method

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1. Introduction

Time-resolved ultrafast electron diffraction (UED) is a powerful diagnostic method which has provided impressive results in transient structures completely with atomic level detail in chemical dynamic processes and the biological functioning processes.[1−6] To probe the ultrafast structural dynamics the temporal width of the probe electron beam has to be shorter than or at least comparable to the time scale of the processes under investigation.[7] Additionally, it must have the capability of capturing the transient structure with a single femtosecond electron pulse preferably. Recently, however, the temporal resolution of UED is limited to several hundred femtoseconds, mainly due to space charge (SC)-induced broadening and other non-space-charge (NSC) broadening effects acting to broaden the electron pulse as it propagates. The SC effect is the self-dispersion due to the Coulomb repulsion between electrons in a pulse, which is most significant when the electron density is high. NSC broadening occurs due to the initial photoelectron energy distributions and different traveling times of electrons along different trajectories. The generation of femtosecond (fs) electron pulses would significantly advance this technique, permitting diffraction imaging of the fastest structural dynamics.

A number of suggestions have been made to improve the temporal resolution of UED and generate sub-100-fs or even shorter electron pulses. These include slicing an ultrashort electron pulse out of a longer one by femtosecond electron pulse gating with surface plasmons[8] or by the concept of tilted optical pulses into diffraction and deflection imaging techniques which allow us to reach limits of time resolution down to regimes of a few femtoseconds and, pos-

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sibly, attoseconds.[9,10] Furthermore, another suggestion is sub-fs pulse generation by electron bunching using a 'temporal lens'[11] or a cavity with a curve-shaped wall,[12] and the simulation of time bunching for a pulsed positron beam.[13]

It was shown that initiating the electrons with an fs-laser generates an energy chirp in the electron pulse which leads to temporal focusing of the electrons at some distance of propagation. The dc electric field allows the parameters of the electric field to be chosen in a wide range for optimizing the electron pulse duration at a specified distance from the target. Simulations with 2000 electrons in the bunch yielded a pulse duration of 55 fs. Whereas for single electrons, i.e., for the case without SC and the number of electrons is less than 2000, 'effective' pulse durations in the attosecond range were predicted. An effective pulse duration may be defined as the standard deviation of the arrival time of the electrons averaged over a large number of events.

In this paper, the studies on temporal compressing of photoelectron bunch with non-stationary electromagnetic fields are continued. The time focusing technique utilizes a combination of dc and time-dependent fields, thus dividing the tasks of acceleration of the electrons and generating the chirp required for bunching between two parts of the system. An appropriate chirp of the electron pulse means that the electrons which are leading the bunch have lower energy, thus propagating with slower velocity whereas the electrons trailing the bunch have a higher energy and thus propagate faster.

The result of the computer modeling of the photoelectron gun with combined time-dependent electric field has shown that, owing to the redistribution of the full energy between the particles inside the bunch when traveling through the time-dependent field region, the system proposed ensures both spatial focusing of the bunch and its temporal compressing down to a femtosecond level. The space charge (SC) effects in time-dependent field are not crucial as in the static one, so they can be neglected in our discussion.[11]

2. The time focusing technique

The basic outline of the new electron gun is shown in Fig. 1. Initial acceleration is achieved with a dc gap similar to those used in state-of-the-art photo-guns: a photocathode consisting of a quartz plate with a thin layer of metal (e.g., gold, silver or copper) is illuminated from the rear side by an fs-laser. Either the photon energy of the laser has to be above the work function of the metal (which typically requires the 3rd harmonic of a titanium sapphire laser) or one may use multiphoton photo-emission. The time-dependent fields are generated in the time focusing electrode.

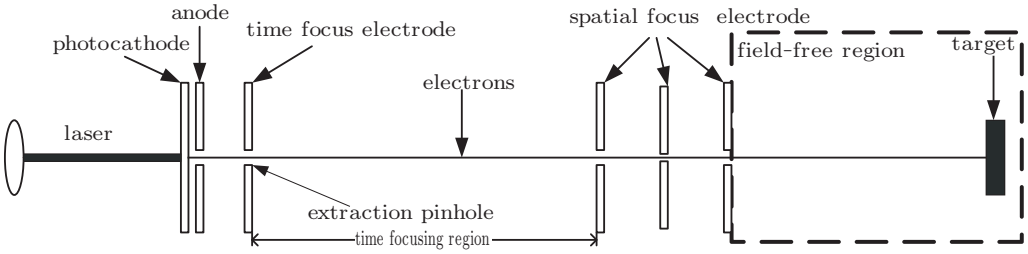


Fig. 1. Basic arrangement of an electron gun using dc and time-dependent field acceleration. Electrons enter and leave the time focusing region through small holes on either side. The region to the target is field-free.

Without considering SC, the basic parameters of the ultra-fast electron gun can be calculated analytically. By solving the nonrelativistic equation of motion one can obtain Δt (temporal advancement at the end of the dc field) on the assumption that $U_{\rm i} \ll U$ in Eq. (1) as compared to those released with zero initial energy,[14]

$$\Delta t(\mathrm{s}) = 3.37 \times 10^{-9} (U_{\rm i})^{1/2} d/U, \qquad (1)$$

where d is the width of the dc gap in millimeters, $U_{\rm i}$ and U are initial energy of electrons released from the cathode and the dc acceleration energy in electron volts (eV) respectively.

The 'advanced' electrons need to delay by a time Δt so that electrons emitted with the same initial energy can reach the target simultaneously. So the velocity change Δu can be given by $\Delta u = u\Delta t/T$, where $T = L/u$ is the time of propagation to the target (L is the transiting distance of the electrons). Finally one

can obtain the Δu.

$$\Delta u = u^2 \Delta t / L. \tag{2}$$

Furthermore, one can obtain ΔU (the required difference in electron energy) by using the nonrelativistic relations $\Delta u = (1/2)\Delta U/U$ and $u = (2eU/m)^{1/2}$.

$$\Delta U(\mathrm{eV}) = \sqrt{8e/m}\frac{\Delta t U^{3/2}}{L}, \tag{3}$$

where e is the elementary charge and m is the electron mass.

3. Simulations of time focusing technique

According to our experience, the initial time of the photoe1ectrons in our calculation is of Gaussian type and sampling in carried out through Monte–Carlo method. The trajectories of electrons were calculated by using the four order Runge–Kutta method. The following parameters were used for the simulation: The electrons are emitted with a truncated Gaussian temporal distribution at the cathode. The laser pulse duration is not a sensitive parameter in these simulations, since it is much shorter than the temporal advancement resulting from the initial velocity spread of the electrons. The dc voltage between the photocathode and anode is 10 kV, and the distance between them is 1 mm, resulting in an electric field as high as 10 kV/mm. The high electric field is used to decrease the chromatic-spherical aberration of the focusing system, and to increase the temporal resolution of the UED system. There is a 3-mm pinhole on the anode to reshape the electron beams and let the electron beams to go through. The initial energy distribution of electrons emitted from the photocathode is taken to be 0.5 eV. The electron emission from the cathode is assumed to be isotropic into the half-space.

First of all, runs were carried out with the slope of the ramp voltage K being 0 V/μs, 8×10^6 V/μs, 8×10^7 V/μs, 4×10^8 V/μs, 8×10^8 V/μs, respectively, and 2000 electrons per pulse with the FWHM (full width at half maximum) of 1 ps without SC taken into account, just as in Fig. 3. The electrons will experience modulating potential field while passing through the time focusing drift region. The speed of photo-electrons will experience a dynamic process while passing through the time focusing region along the axial. The photo-electrons will gain different accelerations with different slopes of the time-dependent voltage. Obviously, while $K < 8\times10^7$ V/μs, the compressing effect of the pulse becomes better and better with increasing K. On the contrary, while $K > 8\times10^7$ V/μs, the compressing effect of the pulse becomes worse and worse with increasing K. $K > 8\times10^7$ V/μs indicates that the initial faster photo-electrons will experience a relatively long period to get to the target, which means the electrons with different longitudinal positions will mix with each other during the transport to the target. The mixing of electrons before getting to the target will destroy the signal imbedded in the photo-electrons from the incident light. This causes the loss of temporal information that is an over-compensation state.[15,16] Here, the photo-electron bunch is not as a carrier of information of the structure of incident optical radiation but as a probing tool for interaction with matter, so we only need to consider the optimum compressing effect.[11]

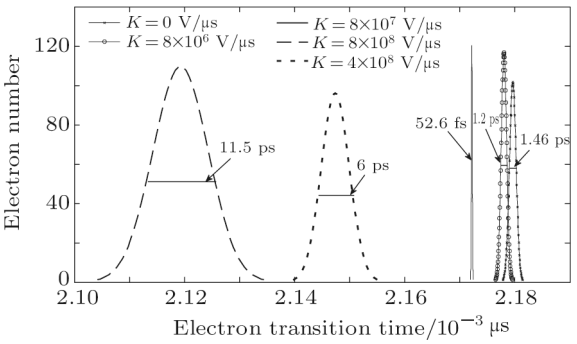


Fig. 2. The pulse distribution at the target with different slopes of the ramp voltage for the same initial electron pulse of a Gaussian fit with FWHM=1 ps.

In displaying the data the question arises how 'pulse duration' of the electron bunch may be defined. It is assumed that the FWHM of a Gaussian fit of the temporal electron bunch distribution is 300 fs. The amplitude of the ramp voltage is linearly changed from 10 V to 4 kV and the slope is 80 V/ps inside the time focusing electrode. As 2000 electrons with time Gaussian distribution enter this region, they will experience a different acceleration voltage due to the relative long traveling time. This is illustrated in Figs. 3(a) and 3(b), which show electron pulse shapes after dc-acceleration at the target as obtained from the simulations. Figure 3(b) also shows a pulse with a width 2σ, where σ is the standard deviation of the electron distribution, defined as the second moment. Since this quantity is directly obtained from the simulations, the 'second-moment pulse duration' is chosen to represent the duration of electron pulses just as follows. Note that for a Gaussian pulse the relation FWHM/σ = 12 applies.

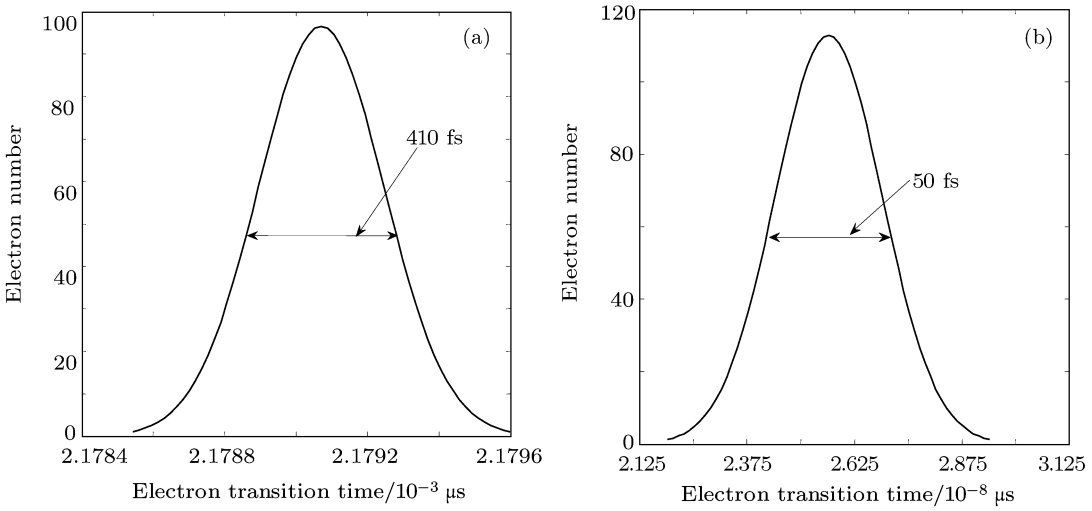


Fig. 3. Electron pulse of 300-fs distribution before (a)/after (b) getting through the time focusing region with the slope of time-dependent voltage added on the time focusing electrode being 8×10^7 V/μs.

It is assumed that the FWHM of a Gaussian fit of the temporal electron bunch distribution is 1 ps and other conditions are the same. The second-moment pulse durations as a function of distance obtained from the simulations are shown in Fig. 4. After the dc acceleration stage the pulse durations acquire σ values of around 600 fs, in fair agreement with the analytical estimate given by Eq. (2). During propagation in the time focusing drift region the pulse duration decreases approximately linearly with distance and reaches a minimum near 105 mm from the cathode, but after passing the time focusing drift region it increases linearly. It is obvious that the minimum pulse duration is 42 fs in this case. As expected, electrons with higher energies are at the leading edge of the bunch since all particles gain the same energy from the static field.

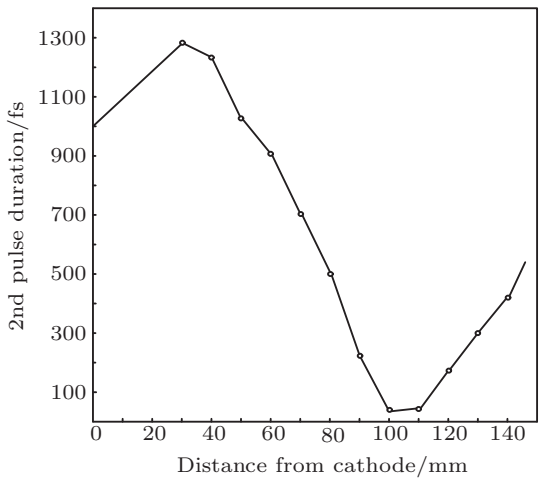


Fig. 4. The pulse duration versus propagation distance of the electrons from the cathode with a dc field being 10 kV/mm.

After passing the time focusing region ∼ 90 mm away from the cathode, the situation is reversed and the higher energy electrons get to the trailing edge of the electron pulse, due to the temporally increasing acceleration field.

4. Stability with respect to parameter variations

The minimum achievable pulse duration is expected to respond sensitively to any changes in the parameters of the accelerating arrangement. In the following section, we scrutinize how small changes in the experimental parameters affect the electron pulse duration.

Firstly, simulations were made to study the effect of dc field between the cathode and the anode on the pulse duration versus propagation distance curve. The anode is always ground electric field. Seen from Fig. 5, the black curve with hollow circle is the previous result from Fig. 4 and a minimum pulse length of (52.6 ± 5) fs for comparison. The dashed line with hollow circle and the black line with solid squares are results with PC=−8 kV and PC=−9 kV respectively. Both curves exhibit longer minimum bunch durations 70 fs and 60 fs at $L = 70$ mm and $L = 80$ mm positions respectively, demonstrating that the temporal elongation is significant. The larger the dc field, the longer the minimum pulse duration is as PC< -10 kV. It should be noted that the minimum pulse duration of the solid line with hollow squares is about 500 fs at $L = 140$ mm, showing that with the increase of the dc

electric field, the temporal elongation becomes more and more infirm, and the position of the minimum pulse is much far away from the cathode. Meanwhile, while PC=−12 kV, the position of the minimum pulse is almost 400 mm far away, which is definitely beyond our requirement. It can be concluded that the electric field determines the position where the electrons lagged behind catch up with the anterior electrons, meanwhile it also acts on the initial energy spread, the space-charge effects and the transverse effects of the electron bunches that limit the final attainable pulse width.

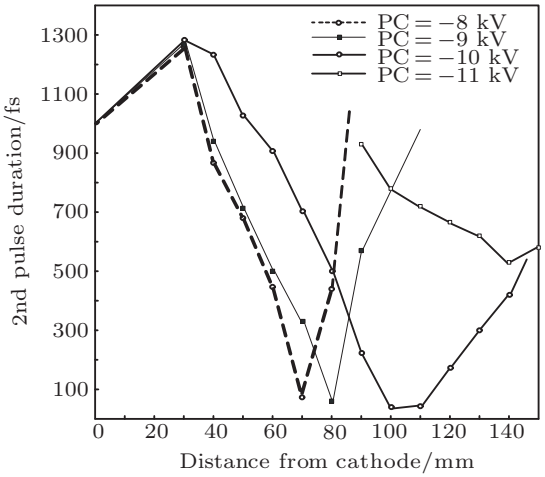


Fig. 5. The pulse duration versus propagation distance of the electrons from the cathode for different dc fields 8 kV/mm, 9 kV/mm, 10 kV/mm, 11 kV/mm.

5. Conclusion

In conclusion, we have presented a new electron gun for direct measurement of the transient structures. The concept is based on accelerating the electrons by means of a dc field and then introducing an appropriate time-dependent field added on the time focusing electrode by means of a time focusing region that leads to temporal focusing after a certain distance of propagation. Through our simulation and calculation, the temporal resolution of the UED can reach 55 fs with 2000 or fewer electrons per pulse in non-space-charge limited regime, which represents a significant step forward in direct determination of transient structures. This ability makes possible the measurement of detailed atomic motion in a unit cell.

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