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Sub-fs electron pulses for ultrafast electron diffraction

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Abstract. We present a new concept for an electron gun generating subrelativistic electron pulses with a duration down to the attosecond range. It is based on a cylindrical RF cavity (a so-called pill-box cavity) oscillating in its TM_{010} eigenmode with a photocathode triggered by a fs-laser pulse. Injecting electrons at an appropriate phase of the RF cycle compensates for their initial velocities and time delays and makes the electrons arrive at a target in a sub-fs temporal window. Such electron pulses will allow nuclear motion and electronic dynamics to be studied on an attosecond time scale.

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Contents

1. Introduction

The advent of few-cycle laser pulses and the generation of attosecond XUV pulses have opened the door to observing new ultrafast phenomena in atoms and molecules [1]–[5]. Such lasers have been used to study molecular dynamics with sub-laser-cycle electron pulses generated by recollision of electrons with the parent atom after optical field ionization [6, 7]. Furthermore, electron orbitals in molecules have been imaged with attosecond time resolution by observation of high harmonics from molecules [8]–[10].

These advances have made attosecond dynamics of electron wave packets and nuclear motion a possible field of study. However, in the aforementioned methods the determination of structure and any changes in it is rather indirect and the evaluation of the data is not straightforward. In contrast to this, ultrafast time-resolved x-ray or electron diffraction make it possible to study changes in the molecule directly by the well-established methods of diffraction analysis [11, 12]. In particular, ultrafast electron diffraction (UED) has become a powerful tool in the investigation of ultrafast processes. Examples of such studies include photodissociation dynamics of molecules, detection of transient dark structures, clarification of bifurcation in the de-excitation of excited molecular states, ring-opening reactions as well as ultrafast surface and bulk dynamics in crystals (see review papers [13, 14]).

State-of-the-art experiments use ultrashort electron pulses generated by illuminating a photocathode with a weak laser pulse and accelerated by a static voltage to energies of a few tens of keV. Unfortunately, however, space-charge broadening still limits the time resolution of this technique to a few hundred fs [15, 16].

On the other hand, it has long been known that accelerating the electrons in an RF field is much more favourable for the generation of ultrashort electron pulses with high brightness than in a DC field (see, for example [17, 18]). More recently, it has been shown that in RF photoguns detrimental effects of nonlinear space charge forces can be eliminated by appropriate shaping of the spatial profile of photoemitted electrons [19, 20]. Moreover, using an ultracold electron source has been proposed to obtain a brightness orders of magnitude higher [21].

Relativistic electron energies are not useful for electron diffraction since such electrons have the disadvantage of a small de Broglie wavelength and a reduced cross-section for elastic scattering. In the present paper, we therefore do not aim at high-brightness relativistic electron pulses, but focus on the generation of electrons at subrelativistic energies with a pulse duration as short as possible. We present a concept which will make it possible to generate electron

pulses with a duration <1 fs, i.e. shorter than the duration of the laser pulse used for emitting the electrons. In this way, the technique of classical electron diffraction can be extended to the attosecond range, allowing one to investigate instantaneous structural changes as well as electronic dynamics in atoms and molecules.

2. Limits of DC-acceleration

A prerequisite of a sub-fs electron gun is that space-charge broadening be prevented. We follow the suggestion put forward in [16] that the pulse charge be reduced to a few electrons or even a single electron per pulse and the repetition rate be correspondingly increased. This requires a laser with MHz repetition rate at energies high enough to generate the electron pulse and excite targets in a pump-probe experiment. Such lasers were recently developed at several laboratories [22]–[26]. Present parameters are a pulse energy of 500 nJ at a repetition rate of 2 MHz and a sub-40 fs pulse duration. Near-term improvements will scale the energy to the μJ level while reducing the pulse duration to <30 fs. 10 fs pulses with microjoule energies will be within reach in the course of the next few years.

Eliminating space-charge broadening increases the temporal resolution by a significant factor. However, new limitations arise, the most important of which results from the initial velocity spread of electrons emitted from a photocathode. Initial electron energies are in the range of 0.25–1 eV if the electrons are released by direct photoemission [27] but can be significantly larger if multiphoton emission is involved [27, 28]. An estimate of the pulse width is derived by solving the nonrelativistic equation of motion of the electrons and is given by the expression [29]

$$\tau_{\mathrm{p}}(\mathrm{s}) = 2.34 \times 10^{-12} (\Delta U)^{1/2} / E_{\mathrm{acc}}, \tag{1}$$

where ΔU is the initial energy spread of the electrons in eV and E_{acc} is the acceleration field in MV m^{-1}. Note that in an electron gun with a static voltage the acceleration field is limited to about 6 MV m^{-1} (= 6 kV mm^{-1}) due to field ionization and vacuum breakdown [30]. As equation (1) shows, it is virtually impossible to generate electron pulses with a duration <100 fs by applying a static acceleration field.

An electron gun using a DC acceleration field was simulated by means of the general particle tracer (GPT) code [31], to be described later. The pulse width is given by a convolution of the laser pulse duration (assumed to be 10 fs) and the broadening resulting from the initial velocity spread of the electrons (see equation (1)). Simulations were carried out at the maximum allowed DC field strength of 6 kV mm^{-1}. The gap width was 6.34 mm and a voltage of 38 kV was assumed. The initial electron temperature was taken to be 1 eV. Under these conditions equation (1) yields a pulse duration of 390 fs. The result of the simulations is shown in figure 1, which displays the pulse duration as a function of the distance from the anode. The analytical estimate is also shown. The simulated pulse duration acquires a length of 375 fs at the anode and then slowly increases to 390 fs at 200 mm of propagation. These results confirm that generating <100 fs pulses with DC fields is not possible even in the absence of space charge.

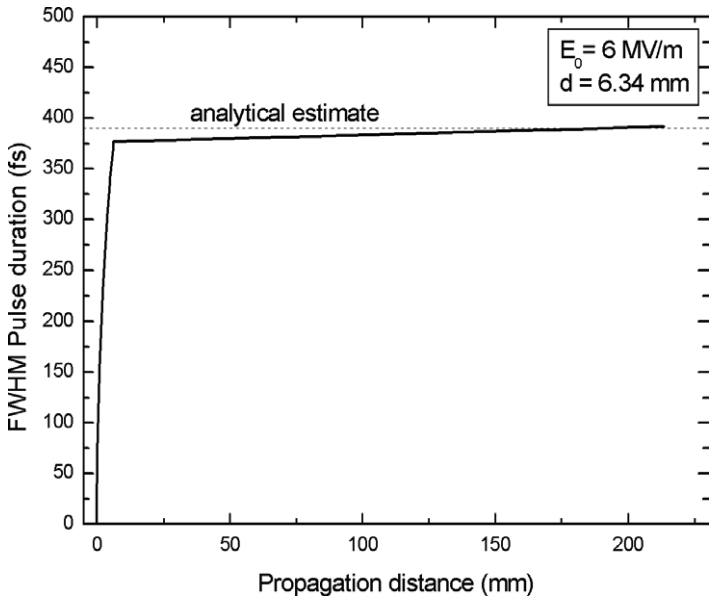

Figure 1. Pulse duration versus propagation distance for a DC electric acceleration field. Space charge effects are switched off. The simulation result and analytical estimate from equation (1) are displayed.

3. The new concept

Here, we present an electron gun design in which an initial electron temperature of 1 eV can be tolerated for generating sub-fs electron pulses. The basic difference is that the acceleration field is applied at RF frequencies rather than with a DC voltage. RF acceleration has two main advantages with respect to DC operation: Firstly, a much higher acceleration field can be applied, in accordance with the so-called Kilpatrick criterion, which can be written in the form [32]

$$f = 1.643\, E^2 \exp(-8.5/E), \tag{2}$$

where f is the frequency in MHz and E is in MV m^{-1}. It should be pointed out that the field given by equation (2) constitutes an operating point rather than a limit.

The second, even more important aspect is that by releasing the electrons at an appropriate phase of the RF cycle the effect of different initial velocities and emission times on the pulse duration can be simultaneously compensated: electrons released with slightly different velocities and time delays experience a slightly different acceleration field and thus acquire a different energy at the end of the acceleration. At a specified distance from the acceleration stage electron trajectories cross each other and an electron pulse with a very short duration is obtained.

The implementation of the new concept relies on the fact that the resonance frequency of a cylindrical cavity oscillating in the TM_{010} mode is independent of its length and depends only on its diameter [33, 34]. Thus, one is relatively free in choosing an acceleration field and a frequency. The resonance frequency of a cylindrical cavity, a so-called pill-box cavity, is given by [33]

$$\nu_{010}(Hz) = 1.147 \times 10^{10}/r_0, \tag{3}$$

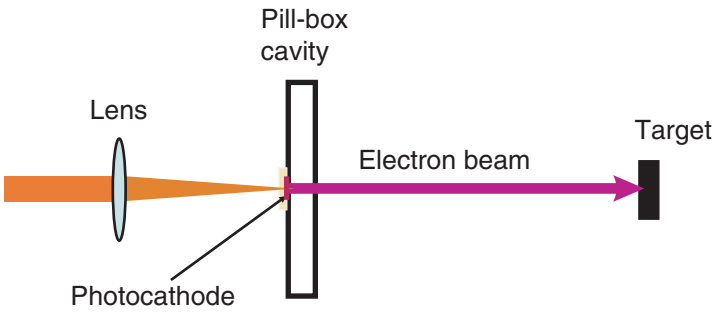

Figure 2. Basic setup of an electron gun using a pill-box cavity. The photocathode consists of a quartz plate coated with a thin layer of a suitable metal. Electrons leave the cavity through a small hole on the right side.

where r_0 is the radius of the cavity in cm. For the TM_{010} mode the electric field in the cavity is longitudinal, whereas the magnetic field is azimuthal. The fields are given by

$$E_z = E_0 J_0\left(\frac{2.405}{r_0} r\right); \quad H_\Theta = H_0 J_1\left(\frac{2.405}{r_0} r\right); \quad H_0 = iE_0/377\Omega \tag{4}$$

In these equations J_0 and J_1 are Bessel functions and r_0 is the radius of the cavity. The relation between E_0 and H_0 applies in SI units. All fields oscillate with a frequency υ_{010}, the magnetic field with a phase of $\pi/2$ with respect to the electric field.

A conceptual drawing of the new electron gun design is shown in figure 2. The photocathode is incorporated in the cavity at the rear side, while the electrons leave through a hole opposite the cathode. The region from the end of the cavity to the target is field-free (see figure 2). The fields given by equation (4) are optimal for particle acceleration: the electric field has a maximum on axis, whereas the magnetic field near the axis increases linearly with radius, resulting in a positive lens effect (see the normalized fields in figure 3).

The photocathode consists of a thin layer (thickness about 50 nm) of a metal such as gold, silver or copper, deposited on a thin quartz substrate. Such a layer is partially transparent to the laser radiation and electrons will therefore be emitted from its entire volume. The layer is also much thinner than the skin depth of the RF radiation (which is about 1 μm) and the electrons will feel the acceleration field throughout the whole layer. For electron emission the photon energy of the laser has to be either above the work function of the metal (which typically requires the 3rd or 4th harmonic of a titanium sapphire laser) or—using a lower photon energy—one may use multiphoton photoemission. We note that the quantum efficiency of this process for a metal is quite low. However, since only one or a few electrons per pulse are required, laser pulse energies in the nanojoule level will be sufficient to drive the photoinjector. Alternatively it would also be possible to reduce the work function of the cathode material by coating it with an alkali metal or to employ a semiconductor for the photocathode.

The delay of the electrons leaving the photocathode will be determined by electron–electron scattering and by the interaction between photoexcited electrons and holes. The first mechanism will lead to a delay which is negligible compared with the duration of the 10 fs laser pulse used in the simulations (see next section). This can be shown by comparing the mean free path of a conduction electron with the layer thickness: The universal curve of the electron mean free

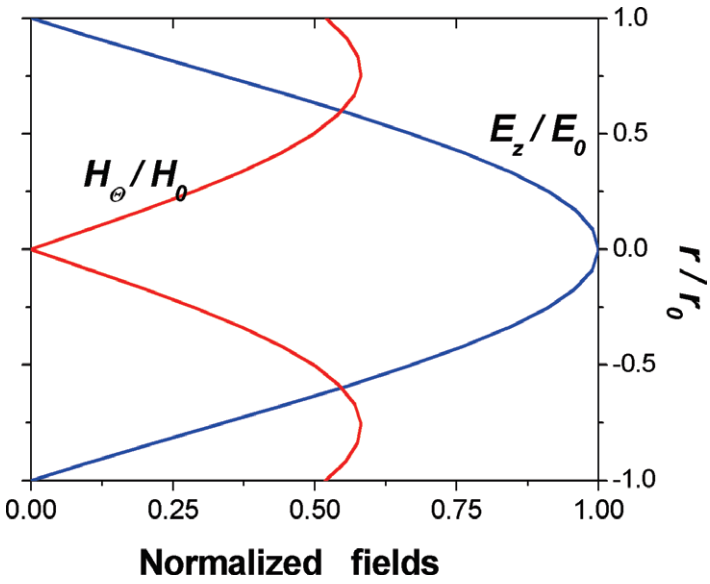

Figure 3. Electric and magnetic fields in a pill-box cavity. All fields are spatially constant along the cavity and oscillate with the TM010 resonant frequency of the cavity given by equation (3).

path (see, for example, [35]) yields a value of about 10 nm at an electron energy of around 5 eV (i.e. well above the work function), which is 20% of the cathode layer thickness. Thus, the electrons will on average experience only a few collisions when leaving the cathode, confirming the assumption of their negligible delay with respect to the laser pulse.

The second mechanism, vice versa interaction of photoexcited electrons and holes, requires further consideration. Experiments using time-resolved two-photon photoemission reveal the existence of image states with lifetimes of 10–20 fs [36]–[38], population of which may lead to delayed photoemission. A delay in the photoemission of electrons from the cathode would be equivalent to a correspondingly longer laser pulse. We note that simulation runs using laser pulses of up to 30 fs duration did not lead to any significant broadening of the electron pulses generated.

4. Simulation results

While the basic physics of the RF photoinjector is rather straightforward, the detailed interaction of the RF field with the particles is quite complicated, especially when the geometry is taken properly into account. This makes it necessary to resort to simulations for calculating the electron pulses for particular parameters. (A one-dimensional quasi-analytical theory was presented in [39].) The pill-box cavity photoinjector was simulated by means of the GPT code [31]. This code is a well-established simulation tool for designing accelerator beam lines. It provides full 3D particle tracking and allows beam line components to be arbitrarily positioned and oriented. Space-charge effects are treated with the model described in [40] but can be switched off to simulate purely ballistic propagation.

The acceleration parameters chosen are an acceleration field of 20 MV m^{-1}, an RF frequency of 5 GHz and a cavity length of 2 mm. At 5 GHz an acceleration field of 20 MV m^{-1} can be

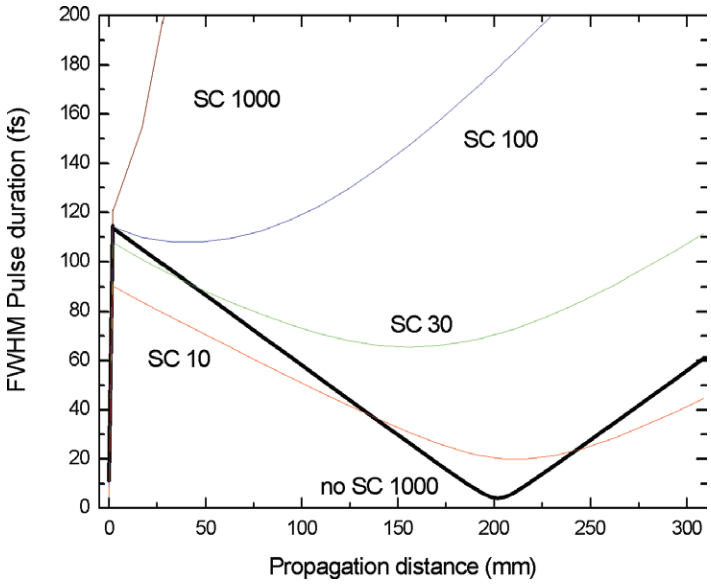

Figure 4. GPT simulation result displaying pulse duration versus propagation length. For cavity parameters see text. The phase is optimized for phase-focusing at 200 mm of propagation. FWHM pulse duration with and without space-charge (SC) is shown. The number after SC gives the number of electrons used in the simulation. For the curve labelled 'no SC 1000' space-charge was switched off but 1000 electrons were used to provide good statistics.

safely applied, in accordance with the Kilpatrick criterion given in equation (2). The spot size of the emitting area was taken to be a circle with a FWHM of 20 μm and a Gaussian transverse distribution. The emission of the cathode was assumed to be isotropic with a temperature of 1 eV. The FWHM laser pulse duration was 10 fs with a Gaussian temporal shape. A sample of 1000 electrons was used to provide good enough statistics, but tests made with up to 10^4 electrons gave identical results. Note that pulse duration in this context (with one electron per pulse) denotes an effective pulse width which is caused by the jitter of the electrons arriving at a target.

The particles are emitted from the cathode at times $-13\,\text{fs} < t < 13\,\text{fs}$ with a truncated Gaussian temporal distribution with a FWHM of 10 fs centred at $t = 0$. They are launched into a field oscillating as $E_0 \sin(\omega t + \varphi)$, where E_0 is the peak electric field in the cavity, $\omega = 2\pi\upsilon_{010}$ is the resonant circular frequency of the cavity, and a phase $\varphi > 0$ ascertains that the field increases while the particle starts propagating through the cavity. Thus, particles emitted with slightly different velocities acquire slightly different final energies. The same applies to particles emitted at slightly different times. Using the optimum phase φ, the electron pulse duration can be minimized. Different optimum phases are obtained for different distances to the target. Note that effects of different velocities and different emission times are simultaneously corrected. This is due to the fact that to first order temporal and chromatic aberrations are proportional to each other and are thus eliminated with the same phase [41].

For the first example the phase of the electric field in the cavity was optimized to get the shortest pulse duration with no space charge after $L = 200$ mm of free space propagation. This optimization yielded a phase $\phi = 0.7674$ rad. The FWHM bunch duration as a function of the propagation distance is plotted in figure 4. The pulse elongates to 110 fs at the cavity

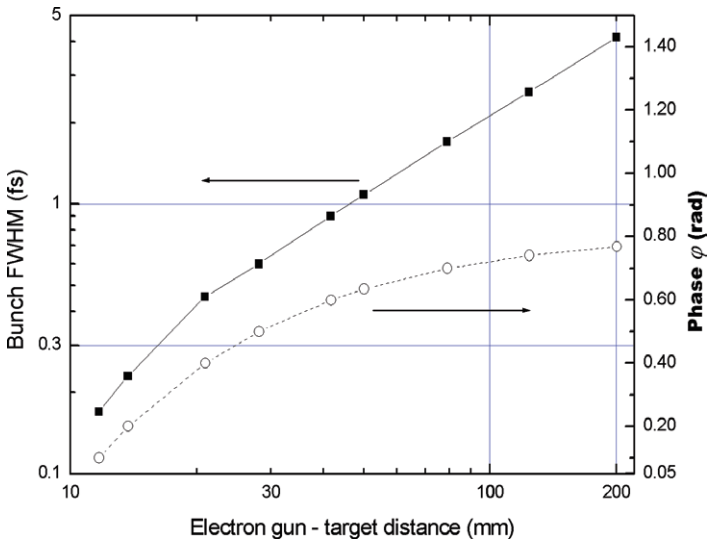

Figure 5. Electron bunch duration for single-electron propagation as a function of cavity–target distance for optimized phases. The optimizing phase is also displayed. For cavity parameters see text.

exit and is then compressed to a minimum duration of 4.2 fs after 200 mm of propagation. The calculated normalized transverse emittance was 6×10^{-3} mm mrad. The figure also displays results obtained with the space charge included. They show that with only 10 electrons the pulse is already broadened to about 20 fs; 30 electrons yielded a minimum pulse duration of 80 fs and with 100 electrons the minimum pulse duration was 110 fs. Note that the minimum pulse durations are obtained at different distances. With 1000 electrons the pulse virtually exploded.

The simulations for single electron propagation show that the shorter the distance to the target the shorter the electron pulse is. An example with the same parameters as previously, but with a distance to target of 50 mm, leads to a pulse duration of 1.1 fs at a phase of 0.634 rad. Reducing the gun–target distance further allows the pulse duration to be reduced down to the attosecond region. Figure 5 displays the pulse duration vs electron gun–target distance at optimized phases. These data are obtained with space-charge effects switched off. The optimizing phase is also plotted into the figure. The shortest pulse duration obtained is 170 as at a distance of 12 mm.

5. Challenges and limitations

Realization of the short electron pulse durations requires the laser pulse to be well synchronized with the RF frequency. This is illustrated in figure 6, which displays the pulse duration as a function of the timing error for 50 and 12 mm distances to target. The figure shows that the pulse duration steeply increases with the timing error from its optimum value. However, even at a timing error of 1 ps the pulse is only lengthened to 7.6 fs for the 12 mm and to 23 fs for the 50 mm distance. To keep the pulse duration below twice the minimum value (i.e. below 340 as and 2.2 fs, respectively) the tolerable timing error is ±32 and ±95 fs. These certainly are challenging figures. We note that locking of a passively mode-locked laser to an RF source with an rms timing jitter of less than 10 fs has been demonstrated [42] and two mode-locked

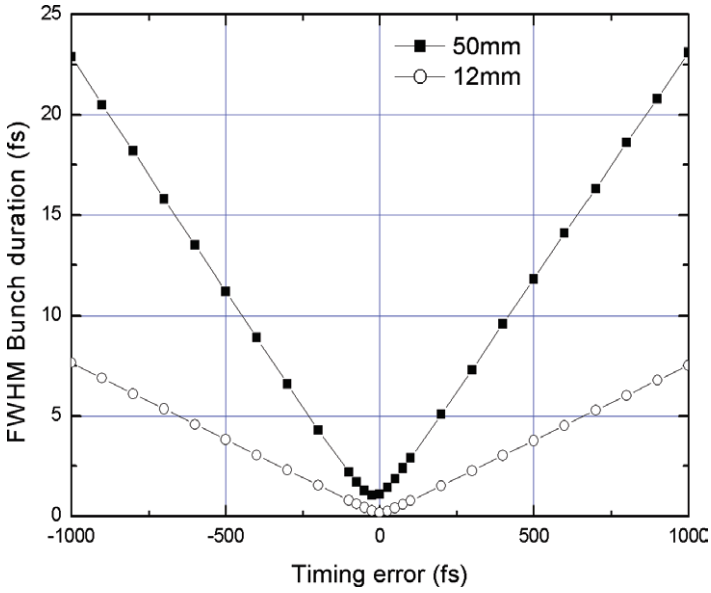

Figure 6. Electron pulse duration versus timing error for $L = 50$ mm and $L = 12$ mm. Space-charge is switched off. The minimum pulse durations at the optimum phases are 1.1 fs and 170 as respectively.

lasers have been synchronized to each other with a timing jitter of less than a femtosecond [43]. However, an additional jitter and a drift may be induced in an RF cavity operated at high power. Synchronization of the laser to an RF signal with a certain level of phase noise will require fast (>50 kHz bandwidth) piezotransducers mounted to the end mirrors of the laser. The availability of such devices gives hope that the figures of ± 32 and ± 95 fs may be achievable even with a high-power RF source.

It is clear from the above that phase focusing has to be paid for by a small energy spread of the electrons which arrive at the target. Figure 7 demonstrates this for three cavity—target distances L by displaying the energy distribution on target of electrons starting with an initial velocity spread corresponding to 1 eV. As can be seen, the resulting energy spread is quite small, reaching approximately 12 eV for the example with $L = 200$ mm, 35 eV for $L = 50$ mm, and 200 eV for $L = 12$ mm. These energy ranges are only a small fraction of the total electron energy, (3×10^{-4} for 200 mm, 9×10^{-4} for 50 mm and 5.7×10^{-3} for 12 mm) and will thus not lead to any significant distortion of an electron diffraction image.

6. Conclusion

In summary we present a new electron gun design which allows sub-fs electron pulses to be generated at energies suitable for ultrafast diffraction studies. The concept is based on accelerating the electrons by an RF field instead of a DC field and by releasing them at an appropriate phase from the photocathode. With electron pulses in the few femtosecond or attosecond time scale the temporal resolution of a pump-probe experiment is no longer determined by the electron pulse duration but by the duration of the laser pulses and their timing jitter. Technical issues which need to be solved include ways of incorporating the photocathode into the pill-box, providing

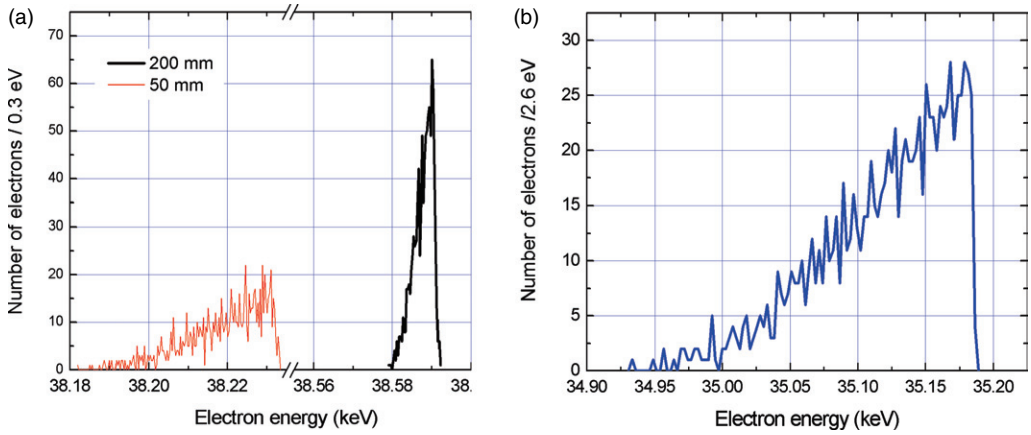

Figure 7. Energy distribution of electrons arriving at target for the case of (a) $L = 200$ mm and $L = 52$ mm and (b) $L = 12$ mm with no space-charge. The initial energy width of the electron distribution is 1 eV. Note axis break in (a) at 38.24 keV. For acceleration parameters see text.

enough power to excite the RF cavity and synchronizing the RF oscillation with the laser pulses. In addition, transverse effects must be taken into account. In a forthcoming publication these problems will be addressed and possible solutions given.

Acknowledgments

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