

# Bunch evolution study in optimization of MeV ultrafast electron diffraction\*

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**Abstract:** Megaelectronvolt ultrafast electron diffraction (UED) is a promising detection tool for ultrafast processes. The quality of diffraction image is determined by the transverse evolution of the probe bunch. In this paper, we study the contributing terms of the emittance and space charge effects to the bunch evolution in the MeV UED scheme, employing a mean-field model with an ellipsoidal distribution as well as particle tracking simulation. The small transverse dimension of the drive laser is found to be critical to improve the reciprocal resolution, exploiting both smaller emittance and larger transverse bunch size before the solenoid. The degradation of the reciprocal spatial resolution caused by the space charge effects should be carefully controlled.

**Key words:** ultrafast electron diffraction, reciprocal spatial resolution, space charge effects, bunch evolution

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## 1 Introduction

With the advancement in research on microcosmic and ultrafast processes, a remarkable demand for new tools for direct visualization arises. Discoveries are made using ultrashort X-ray diffraction in the biology and solid-state physics field [1, 2]. Compared to large X-ray facilities such as the Linac Coherent Light Source (LCLS) [3], ultrafast electron diffraction (UED) is an ideal tool for larger cross-section and tabletop-scale convenience [4–6]. In the pump-probe experiment, an electron bunch of sub picosecond length can be generated and sent to the interested sample to record its structural information with the diffraction pattern. When the sample is pumped by an intense laser pulse, by adjusting the arrival time of the electron bunch with respect to the pumping time, a series of diffraction patterns are obtained and the dynamical process is retrieved.

Currently most UED systems employ a high-voltage static electric field to accelerate the electron bunch to a range of 30 keV to 60 keV. In this scheme, a major hurdle is the dramatic longitudinal expansion caused by significant space charge effects, and the bunch length puts an up limit of temporal resolution of the system [7]. To alleviate the impact of space charge effects, the electron flux is limited to 3000–6000 electrons per bunch, which results in a sub-picosecond resolution at the price of hun-

dreds of repeating pumpings, required by the signal-to-noise ratio (SNR) for resolving the diffraction pattern [5]. One proposed solution is to generate a long bunch containing millions of electrons and then compressed the bunch to a sub-picosecond length at the sample [8–10]. An alternative method circumvents the space charge effects by accelerating the electrons to megaelectronvolt (MeV) energy using a radio frequency (RF) gun [11]. In an accelerating field as high as 60 MV/m, the bunch can be boosted to the relativistic regime in several centimeters before significant longitudinal expansion occurs [12–15]. Benefitting from the development and success of the free electron laser as well as the synchronization system, the RF photocathode gun turns out to be a promising candidate for the UED systems.

The primary concern of the UED system is the ultrashort bunch length, which has been studied by plenary models and simulations [16–18]. These studies focus on the longitudinal evolution of bunch, considering the transverse evolution as a part of the model, if not neglecting it at all. In respect of the experiment, the transverse dimension of the probe bunch is of equivalent importance. The spot size on the detector determines the the reciprocal spatial resolution of the diffraction patterns (see the following text). With poor reciprocal spatial resolution, crucial details of the diffraction pattern will be overwhelmed and lost. In the context of static

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electric-field-based keV UED scheme, simulation studies on pattern displacement and distortion have been conducted [19]. In this paper, we study the transverse evolution of the bunch in the RF gun-based MeV UED scheme with analytical models dedicated to this issue, as well as in simulations methods. It should be noted that the RF gun has been studied by the accelerator community for decades as an intense electron source for subsequent acceleration, but not yet as an independent diagnostic instrument generating a bunch of low charge (few pC) and ultrashort length (sub picosecond). Here, we present a thorough analysis of factors on reciprocal spatial resolution in the context of the MeV UED system based on Tsinghua S-band MeV UED setup, considering the RF gun, the focusing solenoid, and the parameters of the drive laser.

The rest of this paper is organized as follows: Section 2 explains the models of the transverse bunch evolution for the MeV UED system, including the underlying principle; Section 3 presents the results of the simulation and corresponding discussion.

## 2 Models of transverse evolution

A schematic of the MeV UED setup is shown in Fig. 1. The electron bunch is generated by a 1.6 cell S-band RF photocathode gun, which is synchronized with the laser system [20]. A solenoid coil is attached next to the RF gun for restricting the bunch transversely and for emittance compensation. The solenoid is followed by the sample of interest downstream. A detector camera is implemented at the end of the beamline. The positions of elements are presented in Table 1. To accelerate the bunch to a kinetic energy of  $E_k=2.8$  MeV with a small energy spread, a launching phase of 20 degrees and a 60 MV/m peak field at the cathode are chosen. More parameters of operation and probe electrons based on the Tsinghua S-band MeV UED setup can be found in [21].

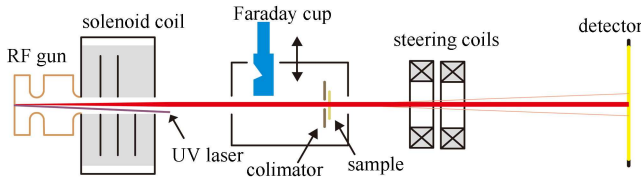


Fig. 1. Schematic of the MeV UED setup.

Table 1. Positions of elements along the beamline.

elements	positions
cathode/cm	0
solenoid/cm	22
sample/cm	73
detector/cm	373

For diffraction patterns, sharper patterns indicate better reciprocal spatial resolution, which can be quantified by the ratio  $\mathcal{R}$  of the diameter  $R$  and the width  $\Delta R$  of the Debye-Scherrer ring [22]. The ring width  $\Delta R$  is determined by the full width at half maximum of the transversal size of the undiffracted beam at the detector, which is 2.35 times of the root mean square (rms) spot size  $\sigma_x$  for a Gaussian distribution [23]. Then, the reciprocal spatial resolution  $\mathcal{R}$  is:

$$\mathcal{R} = \frac{R}{\Delta R} = \frac{2\theta\alpha d}{2.35\sigma_x}, \quad (1)$$

where  $\sigma_x$  is the rms spot size of the bunch at the detector,  $\theta$  is the diffraction angle,  $d$  is the distance from solenoid to detector, and  $\alpha d$  is the distance from sample to detector. According to Eq. (1), the spot size on detector  $\sigma_x$  should be minimized to obtain an optimal reciprocal resolution, since the system setup ( $\alpha d$ ) and characteristics of the sample ( $\theta$ ) are fixed in the experiment.

### 2.1 Model without space charge effects

First, we consider the situation without space charge effects. Using transfer matrix and thin-lens approximation for the solenoid, we can derive the coordinate of the electron at the detector as:

$$\begin{bmatrix} x \\ x' \end{bmatrix} = \begin{bmatrix} 1 & d \\ 0 & 1 \end{bmatrix} \begin{bmatrix} 1 & 0 \\ -1/f & 1 \end{bmatrix} \begin{bmatrix} x_0 \\ x'_0 \end{bmatrix}, \quad (2)$$

where  $d$ ,  $f$ ,  $x$  and  $x'$  denote the distance between the solenoid to the detector, focal length of the solenoid, as well as the position and deviation angle from the axis. The subscript 0 denotes the position before the solenoid. Then the rms value of  $x$  can be derived as:

$$\sigma_x = \sqrt{\sigma_{x_0}^2 \left( \frac{d}{f} - \frac{\sigma_{x_0}^2 + d\sigma_{x_0, x'_0}}{\sigma_{x_0}^2} \right)^2 + \frac{\epsilon_x^2 d^2}{\sigma_{x_0}^2}}, \quad (3)$$

where  $\sigma_x = \sqrt{\langle x^2 \rangle}$ ,  $\sigma_{xx'} = \langle xx' \rangle$  and the emittance  $\epsilon_x = \sqrt{\sigma_{x_0}^2 \sigma_{x'_0}^2 - \sigma_{x_0, x'_0}^2}$ . The  $\langle \rangle$  defines the second central moment of the particle distribution [24]. The minimal value of Eq. (3) can be obtained with proper  $f$  as:

$$\sigma_{x, \min} = \frac{\epsilon_x d}{\sigma_{x_0}}. \quad (4)$$

Then we substitute Eq. (4) into Eq. (1) to obtain the expression of spatial resolution without space charge effect:

$$\mathcal{R}_{\max} = \frac{2\theta\alpha\sigma_{x_0}}{2.35\epsilon_x}. \quad (5)$$

### 2.2 Model with space charge effects

When the space charge effect is significant, the model without Coulomb force underestimates the spot size and the transverse evolution depends on the charge density. In turn, the charge density depends on the dimension

of the bunch, resulting in the coupling of the expansion rates in both dimensions. This close loop feature suggests differential equations are proper for modeling the space charge effects. It should be noted that in the accelerator community where the RF gun is regarded as the source followed by acceleration and focusing, attention are usually focused on the emittance or peak current, instead of the spot size. Here we perform a dedicated analysis for the issue of spot size optimization.

First, we assume the bunch of a uniform three-dimensional ellipsoidal distribution, which maintains the shape of the bunch as well as the uniformity of charge in the linear field [25]. An ellipsoidal bunch can be generated by an intense short laser with “half-circle” radial profile [25]. The blow-out regime [26] is not a widely used regime for a conventional photo injector due to limited current, but the feature of uniform ellipsoidal distribution is favorable for the UED system. We denote the semi-principal axis  $R$  in the transverse direction and the semi-principal axis  $L$  in the longitudinal direction. In the rest frame, the transversal electric field in the ellipse is [25]:

$$E_x = \frac{\rho_0}{\varepsilon_0} M_x x, \quad (6)$$

$$M_x = \frac{1}{2} \left[ 1 - \frac{1+\Gamma^2}{\Gamma^3} (\Gamma - \arctan \Gamma) \right], \quad (7)$$

where  $\rho_0$  is the charge density,  $\varepsilon_0$  is the permittivity of vacuum, and  $\Gamma = \sqrt{R^2/L^2 - 1}$  is the eccentricity. We derive the equations of motion for the particles at the transverse and longitudinal edges of the ellipse [with coordinates of  $(R, 0, 0)$  and  $(0, 0, L)$ ]. Then, we obtain the evolution of the transverse semi-principal axis:

$$\frac{d^2 R}{dt^2} = \frac{4QeM_x}{3\pi RL\varepsilon_0 m}, \quad (8)$$

where  $Q$  is the charge of the bunch,  $e$  is the charge of electron, and  $m$  is the mass of the electron. Then, we transform the coordinate  $(x, y, z, t)$  from the rest frame of the bunch to the laboratory frame with  $R' = R$ ,  $t' = \gamma t$ ,  $L' = L/\gamma$  and  $d^2 z'/dt'^2 = \beta^2 c^2$ . The result is:

$$\frac{d^2 R'}{dz'^2} = \frac{4Qe^2 M'_x}{3\pi R' L' \varepsilon_0 m \beta^2 \gamma^3 c^2}, \quad (9)$$

where  $\gamma$  is the Lorentz factor,  $c$  is the velocity of light in vacuum, and  $\beta$  is the velocity of the bunch relative to the speed of light  $c$ . Similarly, we obtain the equation of motion for the longitudinal direction:

$$\frac{d^2 L'}{dz'^2} = \frac{4Qe^2 M'_z}{3\pi R'^2 \varepsilon_0 m \beta^2 \gamma^3 c^2}. \quad (10)$$

When excluding the effect of space charge, we use the equation of transverse motion with magnetic field  $B_z$  of

the solenoid and emittance  $\epsilon_x$  [27] as:

$$\frac{d^2 \sigma_x}{dz^2} + k_0^2 \sigma_x - \frac{\epsilon_x^2}{\sigma_x^3} = 0, \quad (11)$$

$$k_0 = \frac{eB_z}{2\gamma m \beta c}. \quad (12)$$

For the uniform ellipsoidal charge distribution, the relation between the semi-principal axis and rms value is  $\sigma_x = R/\sqrt{5}$ . Then, we add the space charge term of Eqs. (9) to (11) and obtain the radial differential equation for the ellipsoidal bunch (we omit “ ’ ” here for the laboratory frame):

$$\frac{d^2 R}{dz^2} = k_0^2 R - \frac{25\epsilon_x^2}{R^3} + \frac{4Qe^2 M_x}{3\pi RL\varepsilon_0 m \beta^2 \gamma^3 c^2}. \quad (13)$$

Combining Eqs. (10) and (13), we can investigate the evolution of the transverse size of the bunch quantitatively and evaluate the space charge effects under a specified charge. The result is discussed in Section 3.

### 2.3 Simulation code

We use the particle tracking code ASTRA [28] for simulation purposes. ASTRA is a particle-in-cell code for photoinjector simulation, where different initial distributions of the bunch on the photocathode can be specified. In the simulation, the space charge effects can be switched off for comparison purposes. Using ASTRA, we can obtain bunch parameters such as  $\sigma_x$ ,  $L$ , and expansion rates at the exit of the RF gun. These parameters can be used as initial values for the transfer matrix model without space charge effects or the differential model with space charge effects. We can also track the bunch dimensions from start to end to check the validity of the models above.

## 3 Results and discussion

### 3.1 Model without space charge effects

First we generate bunch distributions of different thermal emittances at the cathode and switch off the space charge effects in ASTRA. Given the absence of the space charge effects and the weak nonlinear field of the RF gun for a sub-picosecond bunch, the emittance is preserved from the cathode [29]. For each specified emittance, we conduct solenoid scanning and obtain the minimized  $\sigma_x$ . In the transfer matrix model, we choose the middle of the solenoid as the thin-lens plane and use the spot size  $\sigma_{x_0}$  from the simulation as the input values. The results of the model and simulation are compared in Fig. 2.

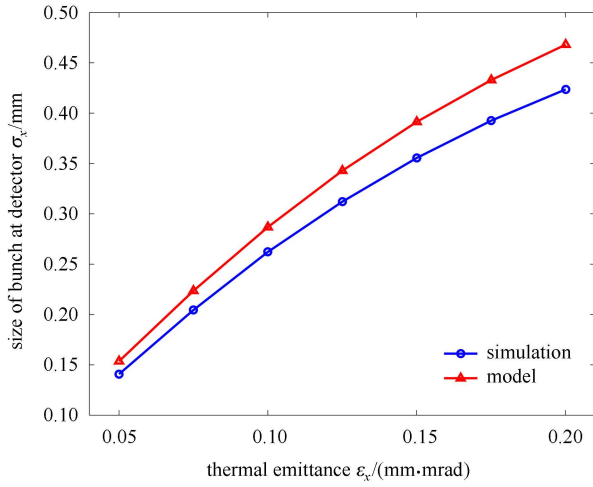


Fig. 2. Optimized transverse size of the bunch at the detector determined by transfer matrix model and ASTRA result without space charge effect.

The results have similar trends and the discrepancy between both results is within 10%. As indicated by Eq. (4), the transverse size on the detector  $\sigma_x$  decreases with emittance  $\epsilon_x$ . The small discrepancy between the model and the simulation may be attributed to the effective distance  $d$  and the thin-lens approximation. Using this model, we can also estimate the influence of energy spread on  $\sigma_x$ . Firstly, the focal length of the solenoid can be derived as:

$$f = \frac{1}{\int (eB_z/2\beta\gamma mc)^2 dz}. \quad (14)$$

Then the smearing of ring caused by the energy spread can be calculated as:

$$\frac{\Delta x}{x} = -\frac{\Delta f}{f} = -\frac{2\gamma\Delta\gamma}{\gamma^2-1}. \quad (15)$$

For a typical relative energy spread ( $\Delta\gamma/\gamma$ ) smaller than 1% for the MeV UED system, the effect of the energy spread on  $\sigma_x$  can be neglected.

### 3.2 Model with space charge effects

The mean-field model has been proposed and proved effective and efficient for modeling the evolution of the bunch length and energy spread, in the keV UED regime [16–18]. In the mean-field model, the bunch is modeled as a cylinder with an evolving aspect ratio smaller than 1 (a thin disk) in the region before the sample. These studies focus on the dramatic longitudinal expansion before the sample as the first concern. However, for the MeV UED regime, the longitudinal expansion is mitigated by the relativistic energy thus not as severe as in the keV UED regime. On the other hand, the transverse expansion in the much longer drifting distance (several meters in the MeV system compared with several cm in

the keV system) results in a significant degradation of the reciprocal spatial resolution. The significant influence of transverse evolution should be taken into consideration of the model construction.

In the ellipsoidal-distribution-based model introduced above, firstly we choose initial values of  $(R, dR/dz)$  and  $(L, dL/dz)$  from the simulation result of ASTRA at  $z=0.5$  m, corresponding to the exit of the solenoid. In this case, we examine the performance of the model in free space. Furthermore, we select another start point upstream at  $z=0.1$  m corresponding to the exit of the gun, including the solenoid scanning in the model. The results of both starting points are presented in Fig. 3, compared with the ASTRA simulation. The consistency between the model and simulation is excellent for a region either with or without the magnetic field.

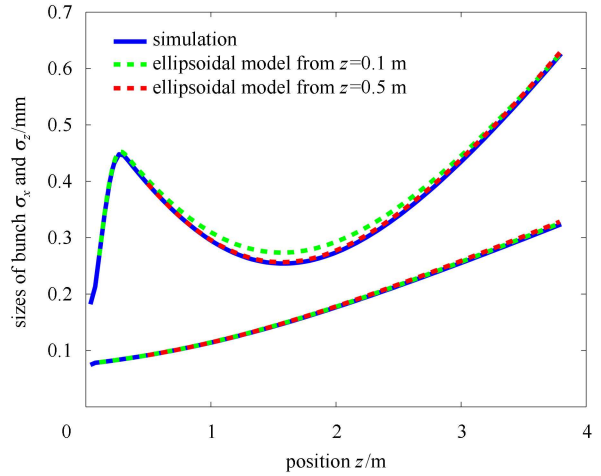


Fig. 3. Transverse (top) and longitudinal (bottom) evolutions of the bunch for three cases: simulation (starting from  $z=0$  m), model from the exit of RF gun (starting from  $z=0.1$  m), and model from the exit of solenoid (starting from  $z=0.5$  m).

With the help of the differential model, we evaluate the space charge effects by examining the extent of transverse expansion for different charge. We choose the exit of the RF gun as the start point and obtain the minimal spot size under different charges. The result (shown in Fig. 4) indicates that the expansion caused by the space charge effect is almost linear with respect to charge. In the case of  $Q=1$  pC, the optimized spot size is four times larger than that in the case of  $Q=0$  due to space charge effects, under an identical initial distribution. While limiting  $Q$  is required to improve the temporal resolution in the keV UED system [16], here we draw the conclusion that to improve the performance of the MeV UED system, especially in the respect of spatial reciprocal resolution, transverse expansion due to the space charge effect must be carefully handled. Otherwise, this issue will

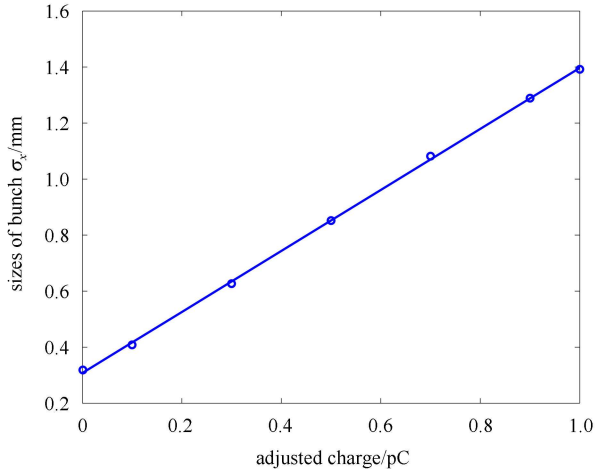


Fig. 4. Optimized transverse size and the linear fitting of different charges calculated by differential model.

hinder the hard-earned advantage of the favorable SNR feature of the MeV UED system.

### 3.3 Optimization of laser spot size

Besides the optimization of the drifting region discussed above, the parameters of the driving laser are also critical. One consideration of the probe bunch is the emittance, consisting of the thermal emittance and the emittance growth [29]. The thermal emittance is proportional to the rms laser spot size  $\sigma_{\text{laser}}$  on the cathode, and takes up a significant part in the low charge regime, like the UED system. Meanwhile the intensity of laser influences the charge density in the drifting region, thus the bunch size evolution, which will be discussed in the following part. Since the dynamics due to the nonlinearity of the RF field is beyond the capability of the models, we study this issue relying on the particle tracking code, from the cathode to the detector. The result of an optimized spot size with respect to the laser spot size is shown in Fig. 5. Other parameters of the laser are listed in Table 2.

Table 2. Simulation parameters of the laser on the cathode.

parameter	value
transversal distribution	flattop
laser spot size $\sigma_{x,\text{laser}}$ (rms)/mm	0.1
longitudinal distribution	Gaussian
pulse length $\sigma_{z,\text{laser}}$ (rms)/fs	300
beam charge $Q$ /pC	1
thermal emittance $\gamma\epsilon_x$ /( $\mu\text{m}/\text{mm}$ )	1

The curve of spot size at the screen (red dash line) shows a convex shape with a peak at moderate laser spot size. To illustrate this result, we scrutinize the components of emittance and focusing effect (blue and red solid line respectively in Fig. 5). The extremely small spot size

at the screen in the bottom left corner can be explained by: 1) the large transverse size at the solenoid caused by the high charge density in the gun and 2) the small emittance proportional to the laser spot size. This is inconsistent with Eq. (4), which has emittance  $\epsilon_x$  in the numerator and size  $\sigma_{x_0}$  in the denominator. When considering space charge effects, the effect is favorable due to the lower charge density (indicated by large spot size at solenoid) during drifting. As the laser spot size increases, the emittance term increases monotonously and a turning point of drifting bunch size occurs. The superposition of both effects explains the shape of the spot size at the screen. Notably, the maximum spot size at the screen is lagged behind the minimal spot size at the solenoid. The lag also confirms the influence of the increasing emittance.

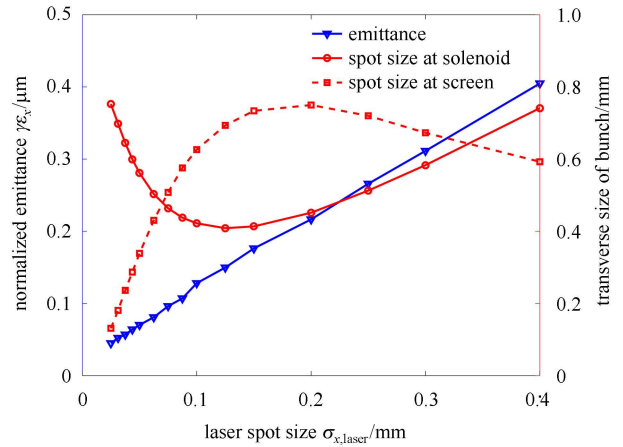


Fig. 5. (color online) Optimized transverse size with respect to the spot size of the laser on the cathode.

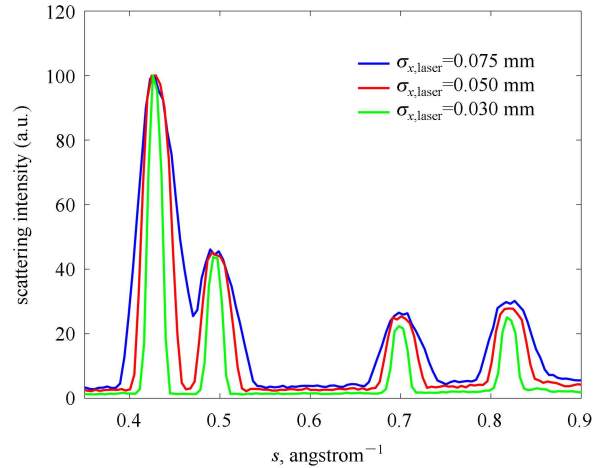


Fig. 6. (color online) Scattering intensity of the diffraction pattern of polycrystalline aluminum with respect to the laser spot size. The momentum transfer  $s$  is expressed as  $s = 2\sin(\theta/2)/\lambda$ , where  $\theta$  is the scattered angle, and  $\lambda$  is the de Broglie wavelength of the electron.

Based on the analysis above, we conduct a thorough simulation for a different laser spot size, including the diffraction process of the polycrystalline aluminum film using the kinematic method. To optimize the resolution, low charge  $Q=0.1$  pC to avoid severe space charge degradation. The result is shown in Fig. 6.

Figure 6 shows that the reciprocal spatial resolution of the diffraction pattern is significantly improved and much sharper than prior simulation results [30] or high quality experiments [23, 31]. For the (111) order of the ring, the optimized reciprocal spatial resolution in the simulation is about  $\mathcal{R}_{\max}=48$ . This value is three times better than the obtained experimental value of about  $\mathcal{R}_{\max} = 15$  for a single shot experimental [31]. Moreover, this value is at the same level as the 20-shot averaged case, which is about  $\mathcal{R}_{\max}=44$  [15]. The resolving ability can be further improved to  $\mathcal{R}_{\max}=78$  by using the charge of 1 pC, relaxing the requirement of temporal resolution.

## 4 Conclusion

In this paper, we focused on the transverse evolution of the probe electron bunch in the MeV UED system to improve the reciprocal spatial resolution. We in-

roduced transfer matrix to model the bunch evolution without space charge effects, the results of which show that the minimal spot size of the bunch at the detector is proportional to the transversal size of the bunch before the solenoid is divided by the emittance. To include the space charge effects, the mean-field model based on the ellipsoidal distribution is modified for characterizing the three dimensional evolutions of the bunch, dedicated for the MeV UED system. This model shows excellent consistency and evaluates the dominating space charge effects in dramatic transverse expansion during the long drifting region. The critical effect of laser spot size on the cathode is also examined. The small emittance and the large transverse size at the solenoid, as well as unmitigated space charge effects, contribute to the extremely small spot size at the screen.

Based on the analysis, we suggest a set of parameters for the experiment to significantly improve the reciprocal spatial resolution. Generally, the models are applicable for different MeV UED setups and the analysis offers a guideline for improving the reciprocal spatial resolution of the MeV UED system.

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