TUNING QUADRUPOLES FOR BRIGHTER AND SHARPER ULTRA-FAST ELECTRON DIFFRACTION IMAGING*

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Abstract

We report our proof-of-principle design and experimental commissioning of broadly tunable and low-cost transverse focusing lens system for MeV-energy electron beams at the ultra-fast electron diffraction (UED) beamline of the Accelerator Test Facility II of BNL. We experimentally demonstrate the independent control over the size and divergence of the electron beam at the sample *via* tunable quadrupoles. By applying online optimization, we achieve minimum beam sizes 75 µm from 1 to 13 pC, two orders of magnitude higher charge density than previously achieved using conventional solenoid technique. Finally, we experimentally demonstrate Bragg-diffraction image (BDI) with significant improvement up to 3 times brighter and 2 times sharper BDI peaks *via* the optimized quadrupoles, improvement larger with higher charge. The result could be crucial for the future single-shot ultra-fast electron microscope development.

INTRODUCTION

Ultra-fast electron diffraction facilities delivering up to 0.8·108 electrons (13 pC) in a single-shot-mode with the electron energy of 3.3 MeV and the temporal resolution of 100 fs to 1 ps [1-8] are providing unique opportunities of simultaneous high temporal and spatial resolution for studies of many processes in physics, chemistry and biology. Among these, resolving the structure of proteins that cannot be crystallized represents one of the most challenging tasks [9]. By employing an accelerator-based radio-frequency (RF) photoinjector as the MeV electron source for the time-resolved electron diffraction, UED takes advantage of strong interaction of electrons with matter and minimizes space charge problems. Due to the almost 1000-fold shorter wavelength of electrons compared to X-rays, UED can resolve much finer structural details enabling us to see how atoms in molecules move and make molecular movies of ultrafast chemical reactions. Therefore, putting both XFEL and UED together will provide a more complete picture in ground breaking studies of all kinds of complex dynamic processes in nature [10].

There are many technical challenges which must be overcome before mega-electron-volt UED can be turned into a significant tool in ultrafast science and technology. Most importantly, a much brighter electron source is required than what is currently available. Conventional round magnetic lenses for MeV-energy electron beams are inherently bulky and expensive due to their focusing power being inversely proportional to the momentum squared [11], preventing them from broad use in scientific community. Quadrupoles are known to have very strong focusing capability, especially for high-energy electron beams due to their focusing strength being inversely proportional to the momentum [12]. A multiplet of quadrupoles, including quadruplet and quintuplet, can form a lens with more desirable properties. In this article, we report our proof-of-principle design and experimental commissioning of broadly tunable and low-cost transverse focusing lens system for MeV-energy electron beams. Such a system based on quadrupole multiplets has been built as a part of the existing instrument for UED experiments at ATF-II, BNL. It has been successfully commissioned with the capability of generating 3.3 MeV electron bunches with 13 pC charge and 1 ps bunch length with the focused beam size 75 µm [1-3]. Transverse beam sizes can be kept constant 75 µm from 1 pC to 13 pC *via* Robust Conjugate Directional Search (RCDS) online optimization [13].

We only focused on improving the intensity and resolution of the diffraction peaks *via* optimizing the quadrupoles in the configuration where the sample was placed upstream of the quadrupoles. The solenoid was used to focus the electron beam on the sample, and the quadrupoles were applied to focus the diffraction image on the detector. This is a kind of proof-of-principle experiment leading to the future ultrafast electron imaging upgrade. Instead of imaging the sample in real space via quadrupole lenses, we focused on providing brighter and sharper diffraction image on the detector using existing quadrupoles.

We have achieved significant improvements in the brightness and sharpness of Bragg-diffraction peaks. Furthermore, it was demonstrated that a single quadrupole fed by a bi-polar power supply can compensate environmental perturbations, such as the remnant magnetic field, x-y asymmetry of the laser spot on cathode, and the misaligned beam trajectory, etc., to achieve the best controllable azimuthal symmetry of diffraction images.

EXPERIMENTAL RESULTS

Beam Based Alignment

Standard beam-based alignment (BBA) technique was applied to determine the relative position of the quadrupole magnetic field center with respect to the nearest beam position monitor, which is a YAG screen. The beam trajectory (blue squares in Fig. 1(a)) relative to quadrupole centers in the vertical direction fits well with a quadratic relation (black curve in Fig. 1(a)). This orbit deviation was caused by the earth magnetic field. After increasing the maximum current of corrector magnet power supplies from 0.1 A to

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* This manuscript has been authored by Brookhaven Science Associates, LLC under Contract No. DE-SC0012704 with the U.S. Department of Energy

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0.2 A, we were able to correct the beam trajectory close to quadrupole centers (blue curve in Fig. 1(b)) with the corrector setting indicated by red squares in Fig. 1(b). This orbit correction made the quadrupole steering effect negligible. The beam trajectory deviating from quadrupole centers in the horizontal direction was much smaller.

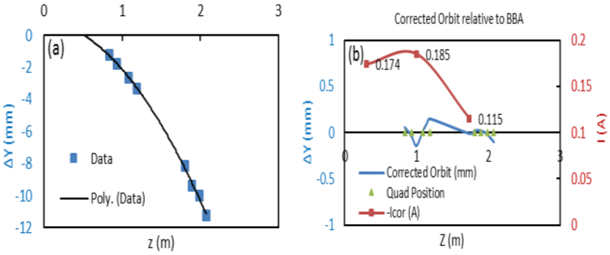

Figure 1(a) (left): Vertical beam trajectory (blue square) with respect to quadrupole centers. 1(b) (right): Corrected vertical beam trajectory (blue curve) with the corrector setting marked as red squares. Quadrupole positions are indicated as green triangles.

BDI Optimization in Imaging Mode

In this configuration, the polycrystalline gold (poly-Au) sample is placed upstream of all quadrupoles. The detail UED beamline layout was presented in our paper [2]. These quadrupole lenses are in-between the sample and the detector, so we call it imaging mode. There is demagnification. The sample is a polycrystalline Au film deposited on a carbon transmission electron microscope (TEM) grid several nanometers thick. The sample is 3mm in diameter, 30nm thick, and has a grain size in the range of tens of nanometers. The grid has a negligible contribution to the electron diffraction.

In imaging mode, there are two independent knobs, the solenoid and the quadrupoles, controlling the charge density at the sample and the focusing of the BDI on the detector. The charge density of the electron beam at the sample can be optimized *via* adjusting the focusing solenoid, and the focusing of the BDI on the detector can be adjusted via tuning the quadrupoles. Focusing the BDI on the detector not only enhances peak intensities and also decreases peak widths therefore achieving a much brighter and sharper image. Without quadrupoles, there is only one knob, i.e., the solenoid. Since the focusing of a BDI is always critically important, we essentially have no control over the charge density at the sample. Hence the use of quadrupoles can significantly improve the BDI qualities. We experimentally compared two cases with and without quadrupoles at the beam charge of 13.9 pC and 1.7 pC.

Figure 2 shows the BD intensity distribution via a 360° averaging at the charge of 13.9 pC. The blue curve represents the case with optimized quadrupoles, having diffraction peaks from the substrate labelled as (sub) and poly-Au diffraction peaks with Miller indexes (111), (200), and (220), etc. The red curve represents the case without quadrupoles, having peaks (sub) and (111), etc. The enhancement of the peak intensity due to applying optimized quadrupoles can be explained as the following:

- the similar peaks, with and without quadrupoles, have different peak positions. We analysed the profile along the horizontal axis between 40 to 110 pixel, which covers the peak with the smallest divergence angle labelled as (sub). This peak is positioned at pixel 52.5 in the blue curve and at pixel 68.5 in the red curve respectively. The position difference is caused by the quadrupole demagnification, about (52.5/68.6) ≈ 0.8;
- the peak width of the blue curve is about 55% of the peak width of the red curve. The narrowing of the peak width is about a factor of (1/0.55) ≈ 1.8. Peak widths were obtained via the detail analysis using Gaussian fitting.
- since we have two independent knobs in the optimized quadrupole case, the solenoid focusing the beam at the sample and quadrupoles focusing the BDI on the detector, the brightness enhancement due to different solenoid settings, with quadrupoles (I_{sol}=129A) and without quadrupoles (I_{sol}=126A), is about 1.3. This enhancement was estimated using the calibration curve in Fig. 3.

The intensity enhancement is the combinatory effect of all three factors listed above. The first one is a factor of 0.8^{-1} increase due to the demagnification. The second one is a factor of 1.8 increase due to the narrowing of the peak width. The third one is a factor of 1.3 increase due to the higher charge density at the sample. Therefore, the enhancement of the peak intensity should be 2.9 agreeing reasonably well with the result obtained by directly analysing the experimental data, about 2.7.

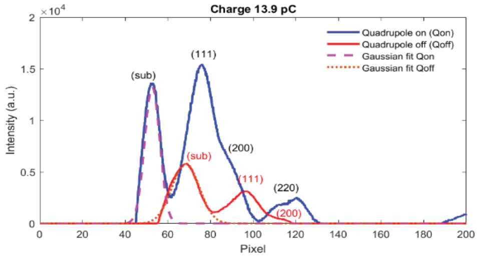

Figure 2: Compare the BD intensity distribution *via* a 360° averaging after the standard background subtraction and ring distortion correction to the image in two cases with (blue curve) and without quadrupoles (red curve) at the beam charge of 13.9 pC.

Here, we assume that the maximum peak intensity is determined by the charge density of the electron beam at the sample. Only the solenoid current is varied, everything else stays the same. Therefore, the measured maximum peak intensity as a function of the solenoid current has been applied as the calibration of the charge density at the sample (Fig. 3).

Comparing the quadrupole focusing effect on the peak intensity of the BDI in the low charge (1.7 pC) and high charge (13.9 pC) cases, the improvement is larger in the high-charge case. The result favours the high charge case when the quadrupole focusing has a much bigger impact. It will be important for the future single-shot UED/UEM development.

ISBN: 978-3-95450-208-0 doi:10.18429/JACoW-IPAC2019-THPMP053

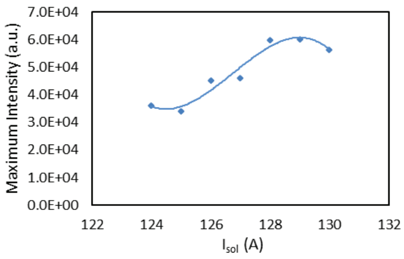

Figure 3: The maximum peak intensity as a function of the solenoid current. The blue curve is the polynomial fit of the experimental data up to the 3rd order.

BDI Distortion Correction via Quadrupole

BDIs are frequently distorted by environmental perturbations. Those perturbations include the remnant magnetic field, the x-y asymmetry of the laser spot on cathode and the misaligned beam trajectory, etc. We experimentally demonstrated that a single quadrupole magnet fed by a bipolar power supply was able to compensate for environmental perturbations and fully restore the azimuthal symmetry of a BDI.

The BDI distortion is described as the roundness error $\epsilon = 2(a-b)/(a+b)$. a and b are the lengths of two principal axises of the ellipse with the highest BDI peak intensity, corresponding to horizontal and vertical axis lengths in the experiment. The roundness error of a BDI from a poly-Au sample was measured as a function of the current of the last quadrupole QF1. As an example, when ΔI_{QF1} = 0.006A, the BDI is shown as Fig. 4(a), and the roundness error was estimated to be 10.0%. Roundness error as a function of the current of QF1 is shown as blue triangles in Fig. 4 (b). The error fits well with a linear relation with the quadrupole current. The error at zero quadrupole current represents a BDI distortion and can be minimized by applying a calculated quadrupole current based on the linear relation shown in Fig. 4(b). Therefore, the x-y symmetry of the BDI on the detector can be restored. Any measurement error caused by environmental perturbations can be well compensated.

Furthermore, in imaging mode, quadrupoles are in-between the sample and the detector. They could potentially introduce skew components, causing the rotation of the BDI on the detector and contributing to the peak broadening of the BD intensity distribution in the 360° averaging data analysis. Those skew errors can be fully compensated by a tunable skew quadrupole.

CONCLUSION

The high-charge, high-brightness, low-energy UED facility has been commissioned at ATF-II, BNL with the capability of generating 3.3 MeV electron bunches up to 13 pC charge (0.8·108 electrons), 75 µm focused transverse beam size, and 1 ps bunch length. The charge density of the electron beam is about two orders of magnitude higher than what has been achieved previously using a solenoid only. Our proof-of-principal experiment has shown that when increasing the electron beam charge from 1 to 13 pC, the space-charge induced growth of the beam size can be compensated by online optimization of the quadrupoles.

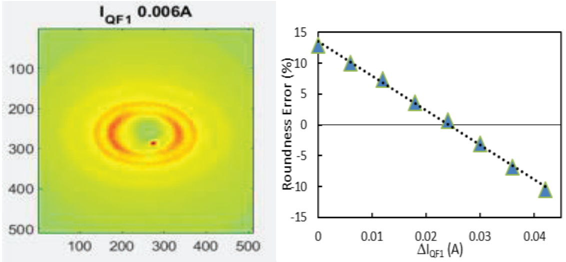

Figure 4(a) (left): BDI when ΔI_{QF1} = 0.006A, and 4(b) (right): The roundness error of BDI as a function of the QF1 current variation.

The imaging mode configuration provides two knobs for optimization: the solenoid and quadrupoles can be applied to focus the diffraction image on the detector. It is a kind of proof-of-principle experiment for the future UEM upgrade. Compared to the conventional solenoid-only focusing, the enhancement of the diffraction peak intensity is about a factor of 1.6 and the narrowing of the peak width is about a factor of 1.3 for the low charge of 1.7 pC. The improvement is more significant for the high charge of 13.9 pC: a factor of 3 improvement in the peak intensity and the narrowing of the peak width by a factor 1.8.

Ultrafast electron microscopy, including diffraction, imaging, and spectroscopy, represents a unique opportunity for understanding structural dynamics and the behaviour of matter under conditions far away from equilibrium at the required time and length scales. A successful quadruple-based transverse focusing system for higher charge density and smaller divergence of the electron beam, as the one we demonstrated here, can play a critical role in advancing the field and make UED and UEM more accessible to a broad scientific community. Compared to x-rays, the UED/UEM system will allow us to take advantage of the unique scattering power of electrons in the presence of space charge to discover emergent properties and dynamical behaviour of exotic material systems.

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