RECENT PROGRESS OF MeV ULTRAFAST ELECTRON DIFFRACTION AT TSINGHUA UNIVERSITY*

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Abstract

Recent years have witnessed rapid advances of MeV ultrafast electron diffraction (UED), in which high quality, ultrashort, MeV electron pulses from a photocathode RF gun are employed as probes for ultrafast structural dynamics. We've built a prototype MeV UED system at the Accelerator Laboratory of Tsinghua University, optimized the the electron pulse parameters as well as hardware performances, and achieved high quality single-shot diffraction patterns. Moreover, MeV UED can be operated in a so-called 'continuously time-resolved (CTR)' mode, in which an RF deflecting cavity streaks the electron pulse thus each diffraction pattern constitutes an 'atomic movie'. We report our experimental progress on MeV UED in this paper.

INTRODUCTION

High brightness, ultrashort electron pulses are indispensable tools that enable us to peer into the ultrasmall and ultrafast world, observing structures with atomic-scale precision and tracing their changes in real time, and eventually understand the underlying physics. Generally, the wavelength has to be roughly equal to or shorter than the inter-atomic spacing (a few ångströms), and the pulse duration is desired to be as short as one atomic vibrational period (1 picosecond to 100 femtoseconds). One approach is to generate high quality X-rays from electron pulses via free-electron lasers (FELs) or Thomson scattering, and the other is to directly use electrons as the structural probe.

In 1980s, G. Mourou and S. Williamson proposed and demonstrated ultrafast electron diffraction (UED) [1], in which ps-long electron pulses are generated via photoemission with ps laser pulses, accelerated by a direct-current (DC) high voltage to tens of keVs, and then probe the sample structures on atomic scales through diffraction. This novel concept and technique allowed for the first time investigations of structural changes on the ps time domain. Since then, it has evolved as a powerful tool for ultrafast structural dynamics studies in chemistry, biological and material sciences, and its capabilities have been steadily improved with the advances of the laser, electron source and detector performances [2, 3].

However, it remains challenging to approach a 100 fs temporal resolving power with DC-based keV UED, mainly due to the strong space charge effects which cause the electron pulse duration to grow rapidly with increasing charge density and time-of-flight. With a most compact DC gun design and as close as 3 cm between the photocathode and the diffraction sample [4], $\sim 10^4$ electrons can be encompassed in a $\sim$350 fs electron pulse, and $\sim$10 such pulses should be accumulated to obtain a high signal-to-noise ratio (SNR) diffraction pattern (DP). Beyond using the conventional DC gun alone, there are recent proposals to employ a radio-frequency (RF) cavity to exert a time-energy correlation (chirp) to the electron pulse thus the pulse durations get compressed by velocity bunching [5, 6]. It's shown in simulation that 100 keV, 0.1 pC electron pulses can be compressed to $\sim$100 fs or shorter.

On the other hand, using photocathode RF guns, the 'brightest' electron sources in the accelerator family, it's not uncommon for an MeV, 100 fs electron pulse to contain $10^6 - 10^7$ electrons. It's first proposed by H. Ihee and X. J. Wang to employ these electron pulses for UED [7, 8]. MeV UED would allow not only higher temporal resolution and single-shot DPs, but also thicker samples to be studied.

Challenges in bringing MeV UED from a concept to a reality include what kind of MeV electron pulse parameters are required to generate high quality DPs and how to realize them, whether the detection efficiency and spatial resolution for MeV electrons are high enough, and whether the RF-laser synchronization jitter deteriorate the temporal resolution even though the high charge electron pulses can be ultrashort [9], etc. Radiation damage would not be a constrain since the electron fluence in MeV UED (10^6 electrons/$\sim$1 mm diameter spot-size) is only 1 electron per 8×10^7 Å^2, roughly 10 orders of magnitude lower than that in the high energy transmission electron microscopy (10^8 electrons/$\sim$100 nm diameter spot-size) [10].

Several groups have made or are now making experimental efforts to demonstrate and improve MeV UED, including the SLAC-Brown University collaboration [11], the Pohang Accelerator Lab at POSTECH [12], the Pegasus Lab at UCLA [13, 14, 15], the Source Development Lab at BNL [16], we at Tsinghua University [17, 18], and the Osaka University-KEK collaboration [19]. We present our recent progress on MeV UED in this paper.

CONVENTIONAL MODE EXPERIMENT

We built a prototype MeV UED system at the Tsinghua Thomson scattering X-ray source facility [20], the schematic of which is shown in Fig. 1. The positions of the main components are listed in Table 1.

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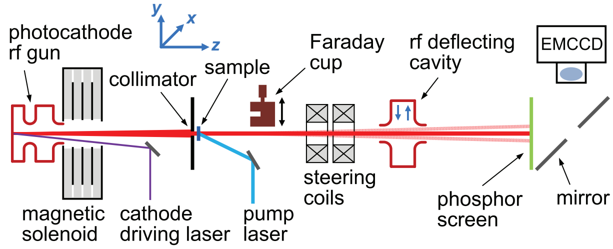

Figure 1: Schematic of the MeV UED system

Table 1: Layout of the MeV UED system

Component	Position (cm)
photocathode	0
magnetic solenoid	21.4
collimator	76.8
diffraction sample	77.0
RF deflecting cavity	166.0
detector screen	377.5

The photocathode RF gun and magnetic solenoid configuration is identical to those used in the injectors for FEL and Thomson scattering facilities, while the electron beam behavior is not necessarily also in the emittance compensation regime. A 1 mm diameter collimator is placed before the diffraction sample. The collimator allows only a small portion of the electron pulse to pass through, thus the beam emittance was dramatically improved at a price of much lower bunch charge.

A typical x–x' phase space distribution after the collimator is shown in Fig. 2. The distribution can be well approximated by a tilted band segment with a uniform root-mean-square (rms) local height of σ_0' and cut off at $\pm D/2$ by the collimation hole. The linearity of this segment stems from the facts that the RF field in the gun, the space charge forces, and the solenoid focusing are all linear in the vicinity of the beam-axis. The negative slope ξ indicates that the beam is focused.

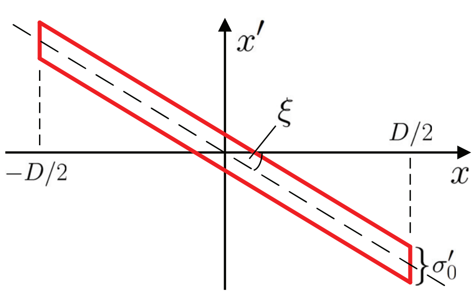

Figure 2: A typical x–x' distribution after collimation

The normalized emittance is roughly $\epsilon_n = \gamma\beta\sigma_0\sigma_0'$, in which σ_0 is the rms spot-size and equals to $D/4$ for a uniform density distribution. With negligible space charge effects and kinetic energy spread, the rms width of each diffraction ring is given by

$$\Delta r = \left[(1+\xi L)^2\sigma_0^2 + L^2\sigma_0'^2\right]^{1/2}, \quad (1)$$

where L is the distance between the sample and the detector screen. With a proper focusing the $1+\xi L$ term can approach 0, thus the ring width is dictated by σ_0'. Smaller σ_0' allows the diffraction rings to be sharper [15, 21]. When the space charge effects are not negligible, we rely on simulation tools to precisely predict the beam behavior.

The quality of the DP can be evaluated by the ratio between the width and radius of the ring $\Delta r/r = \Delta r d_{hkl}/(\lambda L)$, where d_{hkl} is the spacing between the (hkl) lattice plane and λ is the average de Broglie wavelength of the electrons. Equivalently, we can use the rms ring width in reciprocal space $\Delta s = \Delta r/(\lambda L) = d_{hkl}^{-1}\Delta r/r$ to evaluate the DP quality.

A Faraday cup is placed after the collimator to measure the bunch charge with and without collimation. A pair of steering coils are used to correct possible misalignments and fine tune the beam trajectory. The RF deflecting cavity is used to measure the electron bunch length at that location, and we can retrieve the length at sample with aid of simulation codes. The detector consists of a P43 phosphor screen, a 45° first-surface mirror and an Andor iXon$^{\mathrm{EM}}$+ 885 electron-multiplying CCD (EMCCD) camera [22].

With the parameters listed in Table. 2, we obtained a high quality single-shot DP of a ∼200 nm polycrystalline aluminum foil, as shown in Fig. 3.

Table 2: Parameters for the single-shot DP shown in Fig. 3

Parameter	Value	Unit
UV laser pulse duration σ_t^{uv} (rms)	0.8	ps
UV laser spot-size σ_r^{uv} (rms)	0.15	mm
RF field amplitude at the cathode E_a	60	MV/m
electron bunch launching phase ϕ_0	20	Deg
magnetic solenoid strength B_0	0.122	Tesla
collimation hole diameter D	1.0	mm
bunch charge before collimation Q_0	32	pC
bunch charge after collimation Q_s	0.79	pC
kinetic energy at the sample E_k	2.76	MeV
bunch length at the sample σ_t (rms)	2.5	ps

The DP is much improved compared to our previous result [17]. First, thanks to better electron beam qualities, the separations between adjacent rings are more clear and homogeneous, allowing us to trace the evolutions of each ring if there are any structural changes. Secondly, the detection efficiency is much increased. We replaced the previous YAG crystal with a P43 phosphor screen, and used a lens with a larger aperture. We had not calibrated the absolute number of detection efficiency, and are planning to do so as the EM gain calibration for the EMCCD becomes routinely available. Also, magnetic field errors from the steering coils or surrounding components were much reduced, thus the diffraction rings were on longer that obviously elliptical. Using a proper data processing technique [17], we plot the corresponding scattering intensity in Fig. 4, and the width of the diffraction peak is $\Delta s \approx 0.02$ Å^{-1} rms.

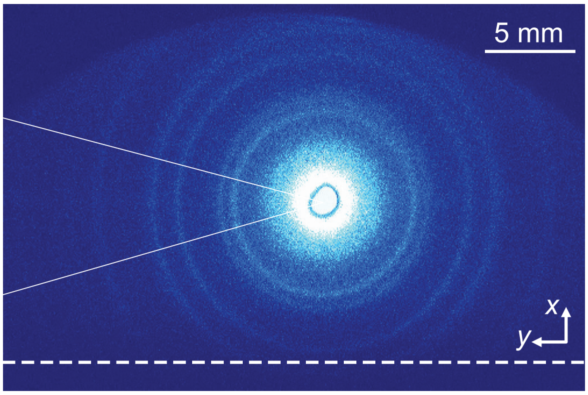

Figure 3: A single-shot MeV UED DP

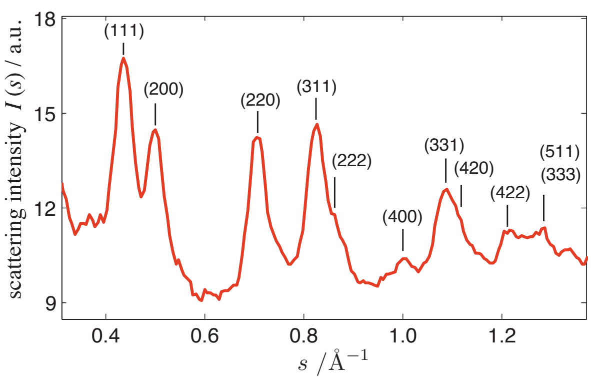

Figure 4: The scattering intensity corresponding to Fig. 3

It's worth noting that with the parameters listed in Table. 2, the longitudinal repulsive space charge forces near photocathode was not negligible, thus the electron beam got negatively chirped (the pulse head had a higher energy) even though the RF chirp should be positive. The longitudinal space charge forces were rapidly damped as the kinetic energies were increased. The chirp was almost frozen after the gun exit however it led to bunch length growth down the beamline. For the conventional mode MeV UED where ultrashort ($\sim$100 fs) bunch length is desired, we may start with low initial charge density to preserve a positive chirp. Simulation results show it's not difficult to achieve 0.1 pC, 100 fs rms electron pulse at the sample with state-of-the-art hardware performances.

CONTINUOUSLY TIME-RESOLVED (CTR) MODE MEV UED

We have reported the results of this novel mode elsewhere [18]. We have obtained single-shot CTR DPs using a static single crystal sample. The demonstrated temporal resolution is $\sim$200 fs. The temporal resolution of this mode is mainly dictated by the resolving power of the RF deflecting cavity rather than the electron bunch length. We are confident that this mode can easily break the 100 fs temporal resolution barrier, however efforts should be made to further improve the efficiency and SNR of the detector.

SUMMARY AND OUTLOOK

We have demonstrated improved electron beam qualities and hardware performances for MeV UED, and are building a dedicated MeV UED facility. Next steps include characterizing the timing between the pump laser and probe electron pulses and further improving the detector, to succeed a time-resolved benchmark experiment thus eventually proving MeV UED a promising and powerful tool.

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REFERENCES

[1] G. Mourou and S. Williamson, Appl. Phys. Lett. **41**, 44 (1982).

[2] M. Chergui and A. H. Zewail, ChemPhysChem **10**, 28 (2009).

[3] J. R. Dwyer *et al.*, Phil. Trans. R. Soc. A **364**, 741 (2006).

[4] G. Sciaini *et al.*, Nature **458**, 56 (2009).

[5] L. Veisz *et al.*, New J. Phys. **9**, 451 (2007).

[6] T. van Oudheusden *et al.*, J. Appl. Phys. **102**, 093501 (2007).

[7] X. J. Wang *et al.*, Proc. PAC03 Conf., pp. 420-422.

[8] X. J. Wang *et al.*, J. Korean Phys. Soc. **48**, 390 (2006).

[9] R. K. Li and C. X. Tang, NIMA **605**, 243 (2009).

[10] A. Takaoka *et al.*, J. Electron Microsc. **46**, 447 (1997).

[11] J. B. Hastings *et al.*, Appl. Phys. Lett. **89**, 184109 (2006).

[12] S. J. Park *et al.*, Proc. 27th FEL Conf., pp. 600-603.

[13] P. Musumeci, J. T. Moody and C. M. Scoby, Ultramicroscopy **108**, 1450 (2008).

[14] P. Musumeci *et al.*, Rev. Sci. Instrum. **80**, 013302 (2009).

[15] P. Musumeci *et al.*, Rev. Sci. Instrum. **81**, 013306 (2010).

[16] Y. Hidaka *et al.*, Proc. PAC09 Conf., TU6PFP018.

[17] R. K. Li *et al.*, Rev. Sci. Instrum. **80**, 083303 (2009).

[18] R. K. Li *et al.*, Rev. Sci. Instrum. **81**, 036110 (2010).

[19] J. F. Yang *et al.*, NIMA, doi:10.1016/j.nima.2010.02.014

[20] C. X. Tang *et al.*, NIMA **608**, S70 (2009).

[21] D. Xiang *et al.*, Proc. PAC05 Conf., pp. 3721-3723.

[22] http://www.andor.com/scientific_cameras/ixon-885/