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MEGA-ELECTRON-VOLT ULTRAFAST ELECTRON DIFFRACTION AND MICROSCOPE AT TSINGHUA UNIVERSITY*

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Abstract

Photocathode gun enabled high brightness, relativistic electron beams with femtoseconds to picosecond tuneable time structure are powerful tools for structural dynamics study. A prototype MeV UED system has been built and operated at Accelerator laboratory of Tsinghua University since a decade ago. Experiments of high quality single-shot static diffraction as well as pump-probe based dynamical process have been successfully conducted, especially the pump-probe experiment in continuously-time resolved mode to achieve better temporal resolution. These studies demonstrated the sub-picosecond timescale and atomic length resolving capabilities of our UED facility. To meet the growing interest of ultrafast sciences based on MeV UED facility, we are upgrading it to a user facility. Meanwhile the concept design of ultrafast electron microscope is proceeding. The current status of UED/UEM facility will be reported in this paper.

INTRODUCTIONS

High brightness electron beams with ultrashort pulse duration are suitable probes for observing structural changes with ultrahigh spatiotemporal resolution. Compared to X-ray probes, electrons serve several unique advantages: 10^4-10^6 times larger scattering cross sections, 10^3 times less radiation damage and much easier manipulability [1]. Moreover, electrons are sensitive to both electrons and nuclei in material, thus making it an ideal complementary tools for understanding ultrafast structural dynamics.

In 1980s, the concept of ultrafast electron diffraction (UED) was proposed and demonstrated by G. Mourou et al [2], based on pump-probe method, where the electrons are generated by high voltage photocathode DC gun, and then probe the laser pumped sample structures on the atomic level and ps time domain. Following this novel and powerful technique, numbers of UED facilities have been constructed and many remarkable experimental results have been generated [3]. However, the temporal resolution of DC-based keV UED facility is limited to picosecond scale because of the strong space charge effect, which causes severe pulse elongating with increasing charge density and drift length. To overcome these barriers, photocathode rf gun was proposed to serve as the high brightness beam source for UED facility [4], in which the electrons are quickly accelerated to a few MeV and space charge force is dramatically suppressed since it scales as γ^{-3}, where γ is the Lorentz factor. Meanwhile, utilization of MeV electron probes also solves the problems of velocity mismatch, where electron travels slower then light in sample, especially in gas sample or surface. Since then, intensive efforts have been devoted to development of MeV UED facilities [5-9], including machine performance, methodology and science application.

On the other hand, diffraction patterns are the Fourier transformation of the nuclei and electrons density distribution of the sample, thus no spatial resolution is achievable is UED. Therefore, to observe directly the image of the sample with sub-angstrom space resolution and sub-ps time resolution is indispensable, which is the main purpose of ultrafast electron microscope (UEM). Currently, most widely used UEM facility are based on photocathode DC gun, where the electron energy is limited to hundreds of keV due to the limited accelerating gradient. Problems of strong space charge effected are also encountered in keV UEM, resulting in pulse duration and energy spread increasing during propagations.

To overcome these problems, a S-band photocathode rf gun based UED prototype has been constructed in Tsinghua university since 2008, both high quality static diffraction and pump-probe experiments were successfully conducted. Now we are upgrading it to a user facility using a more flexible and versatile beamline design and a state-of-out new power source. Meanwhile, the design of UEM is ongoing, where a set of permanent magnet quadruplets (PMQs) are employed as a single imaging unit, different to the commonly used superconducting solenoids. The summary of previous work and the current status of the UED/UEM facility in Tsinghua will be present in this paper.

UED RESEARCH AT THU

The schematic of the prototype UED instrument is shown in Fig.1 [10], where the electrons are generated by a S-band photocathode rf gun and focused by a magnetic solenoid. The collimator located just before the sample is used to select a small portion of electrons to improve diffraction pattern quality. A plug-in Faraday cup is to measure the bunch charge. A pair of steering coils are used to correct possible misalignments and fine tune the beam trajectory. Bunch duration is measured by the deflecting cavity, which also enables the UED system working on the so-called “continuously time resolved (CTR)” mode. The diffraction patterns are captured by the detector system, comprised of a phosphor screen, a 45-deg mirror and an EMCCD perpendicular to the beamline.

A set of machine parameters are optimized by thorough, start-to-end simulations, under the guidance of which a high quality single-shot diffraction pattern of a ~200nm polycrystalline aluminium foil is gained, as shown in Fig.2. It is easy to see that the (111) ring and (200) ring are clearly

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distinguished, indicating a good reciprocal space resolution.

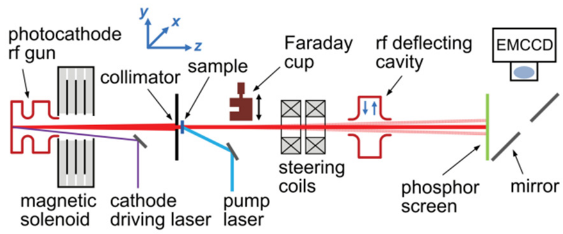

Figure 1: Schematic of the MeV UED system.

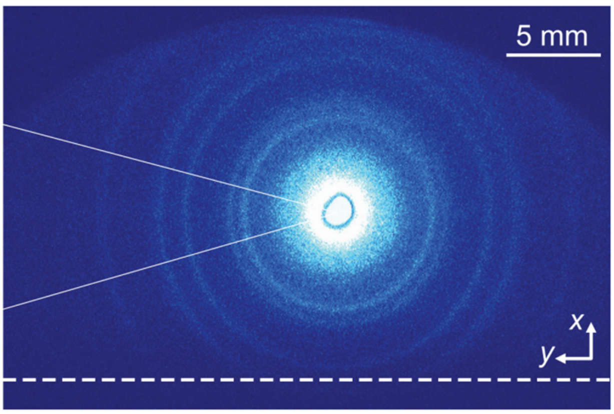

Figure 2: A single-shot MeV UED diffraction pattern.

Apart from static diffraction, pump-probe experiment of single crystal gold sample has been conducted [11]. Typically, the intensity changes of diffraction spot before and after pump ware gained by scanning the time delay between the pump laser and the electron probe. In our experiment, a 10ps long electron pulse is generated and used to probe the sample, then streaked by the deflecting cavity.

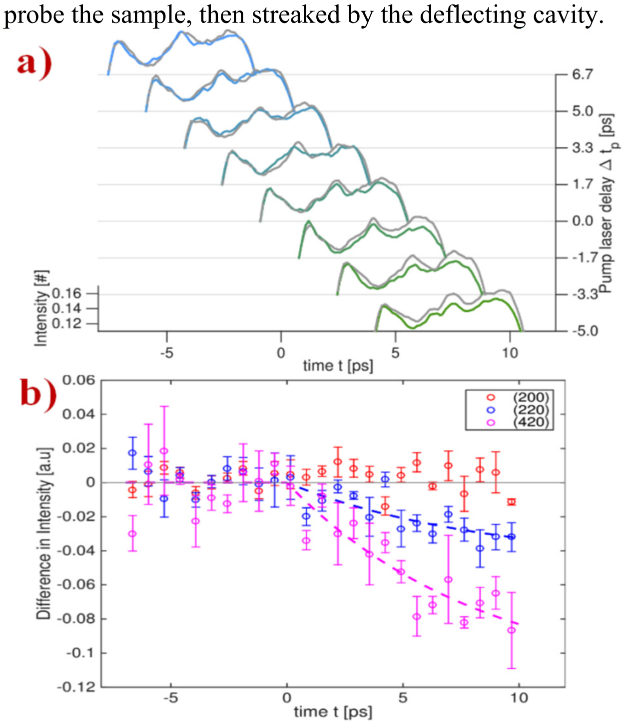

Figure 3: a) intensity of streaked diffraction spot (420) varies with respect to time zero, with pump (colored) and without pump (grey); b) The statistical result of slices of the diffraction patterns at different time delay.

The measured intensity of the streaked (420) diffraction spot at different time delay to pump laser is shown in Fig 3 a), and these in a clear drop in intensity when pump laser is ahead of the electron probe, known as the Debye-Waller effect. The statistical result of slices of the diffraction patterns of (200), (220) and (420) before and after time zero is shown in Fig 3 b). The intensity decay after time zero is fitted with an exponential function of constant of 8.4 ps, which is the Debye-Waller factor.

FUTURE PLAN OF UED/UEM PROJECT

To meet the urgent need of high quality and high stability machine performance for user experiments, we have a newly designed UED/UEM beamline with the following improvement over the prototype:

- A new, state-of-the-art modulator is employed to reduce the amplitude and phase error of the rf system.
- An auxiliary solenoid identical to a C-band photocathode gun solenoid is used to adjust the beamsize on the sample, from a few micrometers to hundreds of micrometers.
- A versatile sample chamber is installed to meet different environment requirement of different user experiments, such as solid sample, gas sample or even liquid sample.

The most significant goal of UED facility is the 100fs time resolution, which is square root of quadratic sum of four items: electron probe length, pump laser length, arrival time error between electrons and lasers at sample and velocity mismatch in sample. Among these factors, electron bunch duration and time-of-arrival jitter are the most concerned ones, since the pump laser is a few to tens of femtoseconds decided by the laser system and velocity mismatch is negligible in thin samples.

In order to get 100fs time resolution, it is necessary to keep the electron bunch length shorter than 100fs. Considering that our current laser shortest duration at 266 nm wavelength is ~127fs, the required bunch length has to be got by lowering the bunch charge, in together with gun compression at low launching phase. We have simulated the bunch length at sample located at 1.5m downstream, as shown in Fig.4.

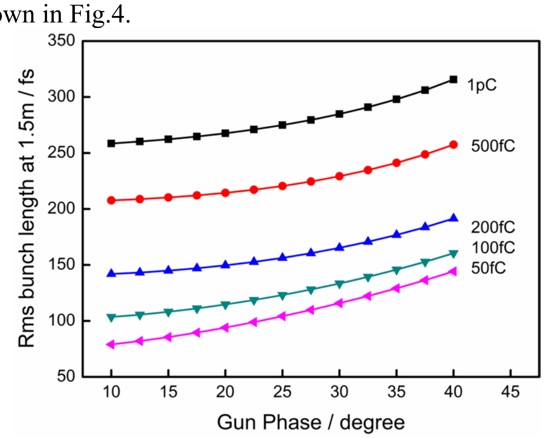

Figure 4: Bunch length vs launching phase.

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It is obvious to see that the bunch charge should be kept below 100 fC, and lower launching phase helps to compress the beam.

The time jitter between the electron probe and pump laser is caused by the amplitude fluctuation of the rf system and laser launching phase error, which includes phase error of the rf system and laser to low level rf (LLRF) phase error. The effect of time of arrival (TOA) jitter with respect to launching phase and rf amplitude jitter is simulated and fitted respectively. Simulation results showed that a degree launching error can cause 670fs TOA jitter, while 0.1% fluctuation of rf amplitude introduces 110fs. Since the laser to LLRF phase error can be controlled between rms 70fs with our current LLRF system, an upgraded modulator with tiny high voltage fluctuation is demanded. With the stability of peak to peak 0.1%, the rf amplitude jitter is about 0.04% and phase error is about rms 40fs. The laser to RF error is then about 80fs and induces a TOA jitter of rms 55fs, while the rf amplitude jitter contributes a TOA jitter of rms 10fs, thus the whole TOA jitter is kept below rms 60 fs. The final time resolution is estimated to be rms 127 fs, which will enable the real-time observation of many ultrafast processes.

As to the design of UEM, permanent magnet quadrupoles (PMQ) are considered as the focusing elements. Typically, solenoids are used as the imaging lens. Reliable and convenient as it is, solenoid coil is often bulky and heavy in MeV UEM. Another alternative option is using PMQ, which is more efficient in focusing high energy electrons than solenoids. A PMQ can offer a high gradient of a few hundreds of T/m with a typical romance of 1.2T~1.4T in material and inner bore of a few millimeters. Recently, a prove-of-principle experiment of using a PMQ based triplet for transmission electron microscope is demonstrated [12]. Since a single PMQ focuses beam in one direction while defocuses in the other, at least two of them need work together to form a focusing element in both x/y directions. Among different configurations, a Russian quadruplet is a promising choice, where the polarities and the strengths of the four quadrupoles are sorted as +A-B+B-A, while the length of the quads(l) and distance (s) between them are l1–s1–l2–s2–l2–s1–l1, thus making this configuration antisymmetric. A single imaging unit based on a Russian quadruplet is designed, the whole transfer matrix of which is:

$$R_{x,y} = \begin{pmatrix} -29.63 & 0 \\ -49.94 & -0.03 \end{pmatrix}. \qquad (1)$$

Where R_{11}=-29.83 is the magnification factor, R_{12}=0 is requirement of point-to-point imaging, the R_{21} element equals -1/f, and the focusing length is then about 2cm.

Parameters of the Russian quadruplet is listed in Table 1. A start-to-end imaging simulation using such a single imaging unit of a virtual sample is present in Fig 5. The beam energy here is 4MeV with rms 0.1% energy spread, while the beam emittance is 100nm and beamsize at sample is 50um.

Table 1: Russian Quadruplet Parameters

Number	Thick	Gradient	Position
Q1	5mm	178.76 T/m	9.9mm
Q2	5mm	-423 T/m	15.9mm
Q3	5mm	423 T/m	22.55mm
Q4	5mm	-178.76 T/m	28.55mm

This clear image proves the reliability of PMQ based imaging system. Cascading of 2~3 imaging units will gain a magnification factor over 1000 times, and then the whole resolution of the UEM is expected to reach tens of nm.

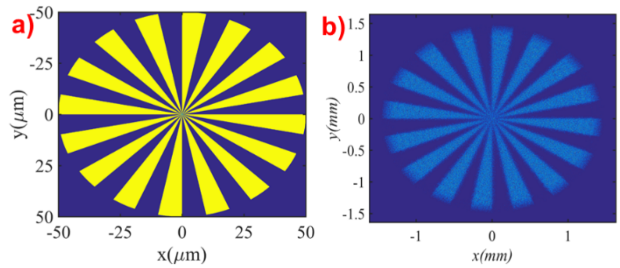

Figure 5: a) distribution of the virtual sample; b) image of a virtual sample with ~30 times magnified.

SUMMARY

Previous work of UED research has laid a solid foundation for the future UED/UEM facility in Tsinghua. The new facility will be installed soon and is expected to serve as a reliable tool for ultrafast sciences.

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