Femtosecond time-resolved MeV electron diffraction

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Abstract

We report the experimental demonstration of femtosecond electron diffraction using high-brightness MeV electron beams. High-quality, single-shot electron diffraction patterns for both polycrystalline aluminum and single-crystal 1T-TaS$_2$ are obtained utilizing a 5 fC ($\sim 3 \times 10^4$ electrons) pulse of electrons at 2.8 MeV. The high quality of the electron diffraction patterns confirms that electron beam has a normalized emittance of $\sim 50$ nm rad. The transverse and longitudinal coherence length is $\sim 11$ and $\sim 2.5$ nm, respectively. The timing jitter between the pump laser and probe electron beam was found to be $\sim 100$ fs (rms). The temporal resolution is demonstrated by observing the evolution of Bragg and superlattice peaks of 1T-TaS$_2$ following an 800 nm optical pump and was found to be 130 fs. Our results demonstrate the advantages of MeV electrons, including large elastic differential scattering cross-section and access to high-order reflections, and the feasibility of ultimately realizing below 10 fs time-resolved electron diffraction.

X-ray free electron laser (FEL) sources and ultrafast electron diffraction (UED) are two of the most powerful tools for exploring the ultrasmall and ultrafast world [1, 2]. The large electron scattering cross section and compactness of electron facilities make UED a particularly attractive option for exploring ultrafast processes and the technique has been used for studying strongly correlated electron systems and revealing transient intermediates in gas phase chemical reactions [3–4]. However, to date, the time resolution of such experiments has been limited by pulse broadening from repulsive space-charge effects (SCE) and the limited acceleration field [5].

To reduce such effects, MeV electron beams generated by a photocathode RF gun have been proposed for UED applications [6–9]. In such schemes, a laser is used to illuminate a photocathode, producing a high-brightness electron beam, and to control the initial spatial and temporal distributions of that beam. An RF cavity then rapidly accelerates the electrons to a few MeV. The RF field also compresses the electron beam as it is accelerated in the time-dependent acceleration field [6]. For electron pulses at MeV energies, the magnetic field induced by the moving electron beam, together with relativistic effects, greatly reduce the SCE. Specifically, the transverse and longitudinal SCE scale as $1/\beta^2 \gamma^3$ and $1/\beta^2 \gamma^2$, respectively [10], where $\beta$ and $\gamma$ are the relativistic velocity and energy, respectively. Thus, increasing the electron energy has the potential to compress more electrons into a shorter electron bunch. Another important advantage of relativistic beams is that they eliminate the velocity mismatch between the pump laser and the electron beam. This mismatch often limits the time resolution of ultrafast dynamics study for gas samples [11, 12]. In addition to the higher temporal resolution, MeV electrons can penetrate thicker samples and have much less dynamic scattering effects. Finally, the higher electron beam energy leads to a larger elastic differential scattering cross-section and a bigger Ewald sphere to fulfill Bragg conditions, allowing access to high-order reflections with good signal-to-noise ratio (SNR) [13].

However, despite significant efforts in MeV-UED in the last few years [9, 14–17], timing jitter between the pump laser and the probing electrons, and the quality of the MeV electron beam, have prevented the ultimate potential of this approach being realized. Here we overcome those challenges and report high-quality, single-
shot electron diffraction patterns for both polycrystalline aluminum and single-crystal 1T-TaS$_2$, each obtained utilizing a 5 fC electron bunch ($\sim 3 \times 10^4$ electrons) at 2.8 MeV. The electron bunch length is estimated to be 40 fs. The timing jitter between the pump laser and the probe electron beam was $\sim 100$ fs (rms) and the overall time resolution, as determined from the evolution of the superlattice peaks of 1T-TaS$_2$ following an 800 nm optical pump, was $\sim 130$ fs (rms). The SNR, sensitivity, and temporal resolution reported here demonstrate a significant improvement over the earlier experiments [9, 14–17]. As a result of these developments, MeV-UED now stands ready to contribute as a powerful probe of ultrafast dynamics, complementing FEL sources with similar sensitivity and temporal characteristics.

To achieve these results, the Brookhaven National Lab (BNL) MeV-UED apparatus reported here was carefully optimized at each step, from electron generation to the detection. Furthermore, there is extensive built-in beam instrumentation which is capable of measuring all the basic transverse and longitudinal electron beam parameters, including charge, beam profile and timing jitter. We have determined previously through simulations that the optimum electron beam energy is between 2 and 4 MeV [15]. Use of higher electron beam energies requires larger RF fields, which not only introduce nonlinear RF effects that degrade the electron beam quality, but also produce significantly more dark current. The operating energy here is optimized at 2.8 MeV.

A schematic layout of the MeV UED system is shown in figure 1. The total footprint of the MeV UED system is about 1.5 by 5 m. The electron source is a BNL-type, 1.6-cell photocathode RF gun [18], capable of producing electron beams with an energy up to 5 MeV. The photocathode is the copper back wall of the RF gun cavity. A UV pulse (4.65 eV, $\sim 150$ fs (FWHM)) from a frequency-tripled Ti:sapphire laser obliquely incident ($\sim 67^\circ$) onto the photocathode is used to generate femtosecond electron beams. The radius of the laser spot on the cathode is $\sim 100$ $\mu$m. The photocathode RF gun is mounted directly on the solenoid magnet to ensure the best alignment between the RF gun and the magnet. The solenoid magnet was optimized for MeV UED, and is followed by a 6-way cross that contains electron beam diagnostics, consisting of a movable Faraday cup and a beam profile monitor. A motorized, 1 mm diameter collimator is also installed on the 6-way cross. The sample chamber is equipped with an $x$-$y$-$\Phi$ motorized manipulator with cryogenic cooling capability. A window on the sample chamber, located 18° off the beam axis, is used to bring the pump laser in, as well as for monitoring the electron beam and pump laser positions. The sample holder can carry up to nine different specimens, plus one YAG screen used for spatial alignment of the pump laser, electron beam and the sample. An RF deflection cavity is installed immediately after the sample chamber. The electron diffraction detector is positioned four meters downstream of the sample chamber to optimize reciprocal space resolution.

One of the challenges in realizing MeV UED has been the diffraction detector. Traditional UED detection techniques, such as a fiber coupler, are not applicable at MeV energies. Furthermore, the x-ray background produced by the MeV electron beam must be carefully avoided. Here, we utilize a 4 cm diameter phosphor screen, positioned perpendicular to the electron beam. The phosphor type, grain size and thickness were all carefully selected to maximize the number of photons generated by the MeV electron beam. A 45° copper mirror with a 5 mm hole is placed behind the phosphor screen. The hole allows the non-diffracted electrons to pass

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**Figure 1.** Schematic of the experimental set-up. UV photons from a Ti:sapphire laser are used to generate electrons in the RF gun. A solenoid magnet focuses the beam onto the detector screen 4 m downstream of the sample. Near-IR pulses from the same Ti:sapphire laser are used to optically pump the sample.
through; a Faraday cup positioned immediately following the copper mirror acts as a beam stop. An Andor electron-multiplying CCD camera with an f#1.4 large-aperture lens records the image reflected by the mirror. The phosphor screen and copper mirror are mounted on an actuator so that they can be retracted and the charge of the all electrons interacting with the sample can be measured accurately.

The UV and pump laser optics are installed on optical tables next to the UED system with a one foot long motorized delay stage on the pump optics arm. The Ti:sapphire laser system consists of an RF synchronized oscillator, a regenerative amplifier and a two-pass amplifier. The oscillator is synchronized to an RF master oscillator at 81.6 MHz, and has a measured time jitter of \( \sim 50 \) fs relative to the RF system when the oscillator environment is controlled to within \( \pm 0.1 \) °F. The equipment is installed at the Source Development Laboratory, BNL [19].

The electron beam is first optimized by adjusting the relative phase between the UV laser and the RF gun, to about 15°. This value was found to optimize the quality of the electron beam and is consistent with simulations and previous experimental results [6]. The electrons were then focused onto the diffraction detector. To reduce the beam size on the sample, a 0.5 or 0.3 mm aperture was placed on the top of the sample.

To demonstrate the quality and time resolution of the MeV UED system, two well-understood samples were tested. The first sample was a 100 nm thick polycrystalline aluminum film; the second, a single-crystal 1T-TaS\(_2\) film with a charge density wave (CDW) modulation. Figures 2(a) and (b) are single-shot, electron diffraction patterns for the two samples each generated by a single 5 fC (3 \( \times \) 10\(^4\) electrons) electron pulse. The previous record for single-shot femtosecond diffraction was with 200 fC pulses on a gold film [20]. Here we have used a factor of 40 fewer electrons in the single pulse. In fact, the increase in sensitivity is even larger than this, when the difference in atomic number (Al versus Au) in the two experiments is taken into consideration. This is another factor of approximately \( (Z_{\text{Au}}/Z_{\text{Al}})^{1.5} = (79/13)^{1.5} \approx 15 \). In Figure 2(b), single-shot diffraction from 1T-TaS\(_2\) is shown. Both the Bragg peaks and the CDW superlattice peaks are clearly visible with good SNR. We believe that this is the first single-shot image showing superlattice peaks with a femtosecond electron beam [21]. Figure 2(c) is a 100-shot accumulation of the 1T-TaS\(_2\) diffraction pattern, demonstrating a significant improvement in quality compared to the previous UED using a dc gun [20]. The visibility of the superlattice peaks demonstrates not only the good transverse electron beam quality, but also the good overall SNR (>400) of the system. The improved SNR of our system also comes from the use of a thicker phosphor screen with large grain sizes allowing electrons generate visible photons more efficiently: a MeV electron deposits about 17 keV energy when passing through a 30 \( \mu \)m thick phosphor screen. Since the phosphor conversion efficiency is \( \sim 20 \) eV per photon, this
yields about 850 photons per electron [22]. Our experimental results unambiguously demonstrate the MeV electron diffraction has a higher sensitivity than conventional keV electron diffraction, especially with easy access to high-order reflections, in contrast to the prevalent view [20].

It is worth emphasizing this last point and clarifying confusions in literature. In figure 3(a) we plot the differential cross section as a function of scattering semi-angle $\theta$ (a) and scattering vector $\sin \theta / \lambda$ (b) for different incident electron energies, calculated based on the multiconfigurational Dirac–Fork algorithm. The green vertical lines mark the corresponding lattice planes of TaS$_2$. The excitation error ($S_g$, distance between the Ewald sphere and reciprocal points) for 50 keV (black dash-line) and 2.8 MeV (red dash-line) is also included in (b). (c)–(e) Calculated electron diffraction patterns of TaS$_2$ (normal state without CDW) for thickness of 70 and 200 nm with 50 keV and 2.8 MeV electron beam. (f) Intensity line scan along the h00 (blue) and hh0 (red) direction with solid-line for 2.8 MeV in (c) and dash-line for 50 keV in (e). For comparison the values of the structure factor squared (the length of the vertical green lines) for the corresponding reflections are included to show the disagreement of the 50 keV diffraction patterns with the kinematic theory. The calculations are based on the multislice method with frozen phonon approach. The effects of inelastic scattering were also taken into account.

Figure 3. The advantages of MeV electron diffraction. (a) and (b) Differential cross section of sulfur (S) as a function of scattering semi-angle $\theta$ (a) and scattering vector $\sin \theta / \lambda$ (b) for different incident electron energies, calculated based on the multiconfigurational Dirac–Fork algorithm. The green vertical lines mark the corresponding lattice planes of TaS$_2$. The excitation error ($S_g$, distance between the Ewald sphere and reciprocal points) for 50 keV (black dash-line) and 2.8 MeV (red dash-line) is also included in (b). (c)–(e) Calculated electron diffraction patterns of TaS$_2$ (normal state without CDW) for thickness of 70 and 200 nm with 50 keV and 2.8 MeV electron beam. (f) Intensity line scan along the h00 (blue) and hh0 (red) direction with solid-line for 2.8 MeV in (c) and dash-line for 50 keV in (e). For comparison the values of the structure factor squared (the length of the vertical green lines) for the corresponding reflections are included to show the disagreement of the 50 keV diffraction patterns with the kinematic theory. The calculations are based on the multislice method with frozen phonon approach. The effects of inelastic scattering were also taken into account.
The RF deflector is powered from the same klystron as the RF gun, there is an inherent synchronization between the two. The RF deflector can be calibrated by monitoring the centroid displacement as the RF deflector phase is adjusted (figure 3(b)). For the S-band (2.856 GHz) RF system employed here, the calibration is 1 RF degree = 0.97 ps ≈ 9 pixels (figure 4(b)). The electron beam bunchlength was characterized by observing its spot size change without and with the deflector on. Due to the resolution limit, we can only conclude that the electron beam bunch length is shorter than 100 fs, which is consistent with our estimation of 40 fs, as seen in the simulations.

The electron beam arrival time jitter can be determined by measuring the spread of the electron beam centroid on the diffraction detector. As a result of positional fluctuations due to the laser pointing stability and RF fluctuations, this spread will have a finite width even if the deflector is turned off. When the deflector is turned on, the width of this spread increases because the timing jitter is now converted into spatial jitter and added to the original positional jitter. The difference in these two distribution widths provides a measurement of the timing jitter. From the data of figure 4(c), we estimate the timing jitter is ∼100 fs (rms) as seen by the broadening of the curves.

To demonstrate the overall temporal resolution of our MeV UED, we performed pump-probe experiments on the CDW material 1T-TaS2. To minimize the temporal degradation due to the non-colinear arrangement of pump and probe we tilted the sample 9° with respect to the electron beam for the measurements. Figure 5(a) shows the temporal evolution of the intensity of the Bragg and superlattice peaks following the 800 nm, 150 fs pump laser pulses. The pump laser power density is ∼1.5 mJ cm⁻². We observed similar behavior to that reported in an earlier study [27–30]. Specifically, the atomic motions that are driven by the optically induced change in the electronic energy distribution result in the suppression of the CDW state. Figure 5(b) shows the temporal behavior of the superlattice peak, together with a fit to a Gaussian experimental resolution convolved with the reported time constant of 300 fs [27] for this material, where we have modeled the sample behavior as a simple exponential decay function. We find the best fit for an experimental time resolution of 130 fs (rms). Since the electron bunch length is short (40 fs), this time resolution is dominated by two sources: the timing jitter between the electron beam and pump laser, and pulse length of the pump laser. With state-of-the-art RF and laser technologies, such as solid-state klystron modulators and a diode pumped laser system, we anticipate that this time resolution could be improved by an order of magnitude, to about 10 fs.

In summary, we report high quality, single-shot femto-second electron diffraction with a 5 fC electron bunch at 2.8 MeV. Our experimental results show that MeV UED not only can achieve very high temporal resolution (here 130 fs), but also has significantly improved overall diffraction intensity compared to the existing methods. The optimized MeV electron beam in our system, including the large transverse coherence length, allows us to record weak superlattice reflections with unprecedented quality. Continued improvement of such a UED system, such as solid-state klystron modulator and a state-of-art laser system, could improve the MeV UED temporal resolution to 10 fs. By reducing the laser spot size (from 100 to 10 μm) and thermal emittance, the transverse coherence length could be as large as 100 nm, this may lead to electron diffraction-based coherent diffractive imaging, or ptychography, to visualize aperiodic nano-objects in real space, including nanocrystals, proteins and molecules, with a desirable time resolution. Thus MeV UED stands ready as a technique to complement FEL x-ray sources in the study of ultrafast structure dynamics for a wide range of systems.
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Figure 5. Characterization of time resolution for the MeV UED (normalized intensities). (a) Dynamics of Bragg and superlattice peaks of 1T-TaS2. (b) The overall time resolution of the instrument is estimated to 130 fs by fitting the superlattice data to an exponential decay.