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Mega-electron-volt Femtosecond Electron Micro-diffraction.

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X-ray Free Electron Laser (XFEL) and ultrafast electron diffraction (UED) are powerful tools to study ultrafast structural dynamics in materials, chemistry, and biological sciences with atomic spatial and temporal resolutions [1-2]. While x-ray is sensitive to the electrons of materials, electron interacts with both electrons and nuclei in matter. In the meantime, Electron provides 10^4-10^6 times larger scattering cross sections and 10^3 times less radiation damage per elastic scattering event. Therefore, nuclear geometry determination by UED provides an ideal complementary tool to study of electronic structure by XFEL.

Visualization of ultrafast structural dynamics requires an instrument to provide a probe beam with pulse duration $\leq$ 100fs, which matches the typical atomic time scale. Conventional UED systems usually implement $\sim$10MV/m dc electric field to produce electrons in the tens of keV energy range. Electron beam at such low energies suffers from the strong repulsive space charge forces so that it is practically impossible to obtain sub-picosecond pulses at the specimen with more than a few thousand particles. On the other hand, a rf photoinjector provides electric field which normally exceeds 100MV/m to rapidly accelerate electron to MeV energy level. Space charge at such high energy range is strongly suppressed such that sub-picosecond pulse duration with higher charge is achievable. The high brightness electron beams generated from a rf photoinjector also make it flexible to focus the electrons into small lateral beam size, which enables study over small sample domain for determination of local crystal structure and defects.

Driven by the strong opportunities for ultrafast science provided by MeV electrons, a MeV UED system was constructed at SLAC [3]. Recent efforts have been devoted to commission a second solenoid condenser lens, which is called micro-focusing solenoid, to focus lateral beam size to a few um while retaining pulse duration of ~100fs. A MeV femtosecond electron micro-diffraction (FEMD) system is then enabled. Figure 1 shows a schematic of the MeV FEMD beam line. 266nm UV laser of 40fs full-width-at-half-maximum (FWHM) pulse duration illuminates the photocathode at an incidence angle of 70° to generate photoelectrons. The high electric field in the rf photocathode rapidly accelerates electron bunches to relativistic energy, where the effect of space charge forces is strongly reduced. The gun solenoid immediately after the rf photocathode adjusts the beam divergence at the exit of the rf photocathode, which effectively adjusts the beam size on the collimator located downstream of the gun solenoid. The pinhole on the collimator is used to select the central part of the beam, which effectively filters beam emittance. The emittance-filtered beam is focused into small probe size at the specimen by the micro-focusing solenoid downstream of the collimator. Diffraction pattern is captured by a downstream EMCCD imaging system.

To realize FEMD, 30fC electron beam was accelerated to 3.7MeV in the rf photocathode and sent downstream. The gun solenoid was set at an integrated magnetic field of 0.025 T·m to spread out the electron beam lateral size at the collimator with a pinhole of 100um diameter, such that the electron beam core with 1.5fC charge was selected. The micro-focusing solenoid was set at an integrated magnetic field of 0.011 T·m to focus the beam on the specimen. From a knife-edge measurement, the root-mean-square (rms) lateral beam size was determined to be $\sigma_{r,rms}$=5um. Normalized beam emittance was measured to be 3.1 nm-rad by a solenoid scan. The rms pulse duration was measured from the ultrafast laser induced structural changes of a 25-nm-thick Bi(111) thin film and determined to be $\sigma_{t,rms} = 109$fs [3].

To demonstrate the power of MeV FEMD in resolving local crystal structure, diffraction patterns from Paraffin ($C_{44}H_{90}$) crystals were probed with electron of different lateral beam sizes. Figure 2a shows a scanning transmission electron microscope image of the Paraffin with 30um crystal size. Figure 2b shows a diffraction pattern with electron lateral beam size $\sigma_{r,rms} = 30$um. In this case, multiple crystals were sampled simultaneously. Figure 2c shows a diffraction pattern with electron rms lateral beam $\sigma_{r,rms} =$ 5um, in which single crystal diffraction features were clearly resolved.

With the exceptional properties in lateral beam size and pulse duration, the MeV FEMD system provides great opportunities for scientific study of local crystal structural dynamics at ultrafast time scale in material, chemistry, and biological sciences.

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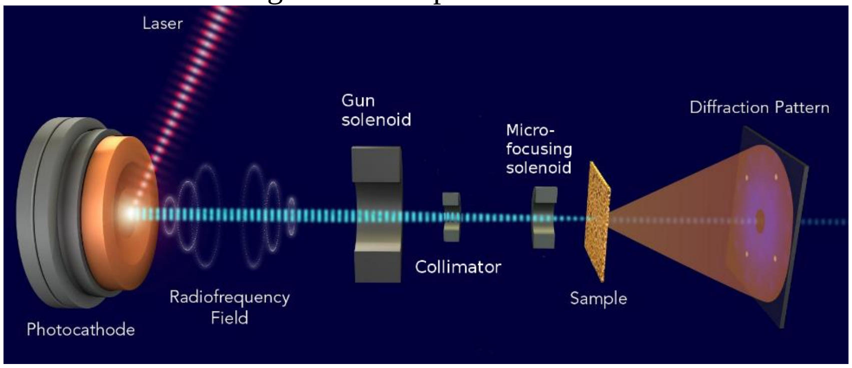

Figure 1. Schematic of the MeV femtosecond electron micro-diffraction beam line (not to scale).

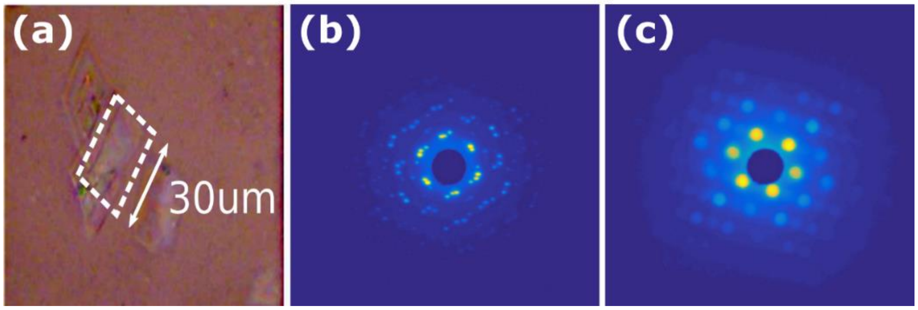

Figure 2. Characterization of MeV FEMD resolving power. (a) TEM image of the Paraffin ($C_{44}H_{90}$) crystals of 30um size under study. (b) Paraffin diffraction pattern sampled by electron beam with $\sigma_{rms} =$ 30um. Multiple crystals were simultaneously sampled by this electron beam. (c) Paraffin diffraction sampled by electron beam with $\sigma_{rms} = 5$um. Single crystal diffraction features were resolved.