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Invited Paper

Ultrafast structural dynamics of materials captured by relativistic electron bunches

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ABSTRACT

Ultrafast electron diffraction (UED) has become a leading technique for investigation of structural dynamics in solids providing high spatial and temporal resolutions. Radio frequency (RF) based photoinjectors providing Mega-electron-volt (MeV) scale electron beams are improving the source brightness and instrument versatility and are largely responsible for advancement of the field of structural dynamics. At Lawrence Berkeley National Laboratory (LBNL), an RF photoinjector gun for ultrafast structural studies using UED has been in development and is now producing high-quality scientific results. Here we describe some factors that enable UED of materials at LBNL and present some exemplary results.

Keywords: UED, Relativistic electron source, ultrafast structural dynamics, HiRES, MeV-UED

1. INTRODUCTION

Studies of ultrafast light-matter interaction in solids can provide critical insights on microscopic processes that govern material properties.[1] Detailed knowledge of how light couples with different degrees of freedom within solid media could open up the prospect to control and manipulate states in these systems and their properties. Various phenomena, such as metal-to-insulator phase transitions,[2] piezoelectric effect[3] and melting can be photoinduced and can lead to drastic changes in electronic structure and rearrangement of atoms.[4] Visualization of structural changes during these processes carries the potential to enrich our understanding of the underlying physics which is a necessary precursor for development of future applications involving these phenomena such as ultrafast solid-state electronic devices[5] and photo-active actuators.[6]

In order to resolve atomic motions during a photoinduced process, few requirements must be met simultaneously: very high spatial ($\sim$ 1Å) and temporal (0.1-1 ps) resolutions must be provided and brightness of the source must be high enough to light up the structural changes.[7] Ultrafast electron diffraction is one of the

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forefront techniques that satisfies these requirements.[8] Along with X-ray free electron lasers (XFELs)[9] —which are complementary tool, UED has been applied to many important problems in condensed matter physics and material sciences, from ultrafast mechanism of non-thermal melting in Al,[10] Au[11] to charge-density wave (CDW) dynamics in two-dimensional (2D) transition metal dichalcogenides[12,13] and flow of energy in metallic films.[14] Like its X-ray counterpart, UED is based on principle of *pump and probe* in which an ultrashort laser pulse initiates the dynamics of interest and time-delayed electron pulses monitor changes in atomic structure by scattering off the interatomic potential and forming diffraction patterns at the detector as illustrated in Figure 1. The information of the structure modulations are encoded in the diffraction images which can be processed and evaluated to extract valuable insights on transient structures.

Ultrafast electron diffraction sources have been in development since 1980s for the pursuit of femtosecond electron pulses for structural dynamics and have seen the emergence of high brightness MeV-scale electron guns across the world such as those at UCLA,[15] SLAC,[16] BNL,[17] and at LBNL.[18] This follows the successful implementation of compact direct current (DC) electron guns[19–23] which however operate at kilo-electron-volt (keV) energies.

In this proceeding, we outline factors that make structural dynamics studies of materials using UED at LBNL possible. We discuss advantages of electrons as structural probes over X-rays and the benefits of relativistic electron beams which are employed in LBNL UED apparatus and is enabling new opportunities for dynamic studies of materials.

2. X-RAYS VS ELECTRONS

X-ray and electrons are complementary tools for studies of structural dynamics, however electrons have some advantages over X-rays which make them very attractive probes for investigation of ultrafast process in solids: they have a factor of 10^6 higher cross-section for elastic scattering and a very favorable elastic to inelastic scattering ratio when compared with X-ray, which means they are particularly suited to study poorly scattering systems, such as organic conductors, provide high contrast and inflict lower sample damage due to the lower dose needed to form diffraction patterns with sufficient signal-to-noise (S/N) ratio for inversion.[24] Electrons also have much shorter wavelength than X-rays and can be manipulated by electron optics which makes them a more promising tool for imaging applications than X-rays.[25]

A potential drawback of using electrons is their charged nature which can lead to significant space charge effects and degradation of beam transverse and longitudinal coherence.[26] However, as will be discussed below, this can be circumvented by application of relativistic electron sources.

3. RELATIVISTIC ELECTRON DIFFRACTION AT LAWRENCE BERKELEY NATIONAL LABORATORY

Relativistic electron beams for ultrafast scattering experiments are typically produced by state-of-the-art RF photoinjectors that are able to achieve field gradients on the order up to 100 MV m^{-1}.[27] A major advantage of relativistic electron beams over non-relativistic beams produced by keV-scale UED setups is the effective suppression of space charge defocusing forces which scales as $\frac{1}{\gamma^2}$, where γ is the Lorentz factor. Therefore, at higher energies of electrons substantial suppression of the space charge occurs.[28] Negating space charge effects drastically improves the beam properties and preserves the brightness of the beam. Shorter pulses (< 0.2 ps) with flux reaching the single-shot limit (10^6 electrons/pulse) are possible with relativistic electrons opening up the possibility to study irreversible processes such as radiation-induced damage in materials and melting.[29] Finally, the very short wavelength associated with high-energy relativistic electrons results in an extremely flat Ewald sphere that intercepts many reciprocal lattice points in crystalline systems fulfilling the Bragg condition and contributes to intensities of higher order diffraction spots.

Ultrafast electron diffraction setup (HiRES: High Repetition-rate Electron Scattering) at LBNL is based on RF photoinjector APEX gun[30] that has been developed for structural dynamics of materials and gas phase targets. Typical parameters of the HiRES beamline, schematic of which is provided in Fig 1b, are summarized in Table 1. The key components of the beamline relating to electron beam generation and its control are a continuous wave (CW) RF gun, solenoids and quadrupole magnets for beam focusing and shaping, dipoles,

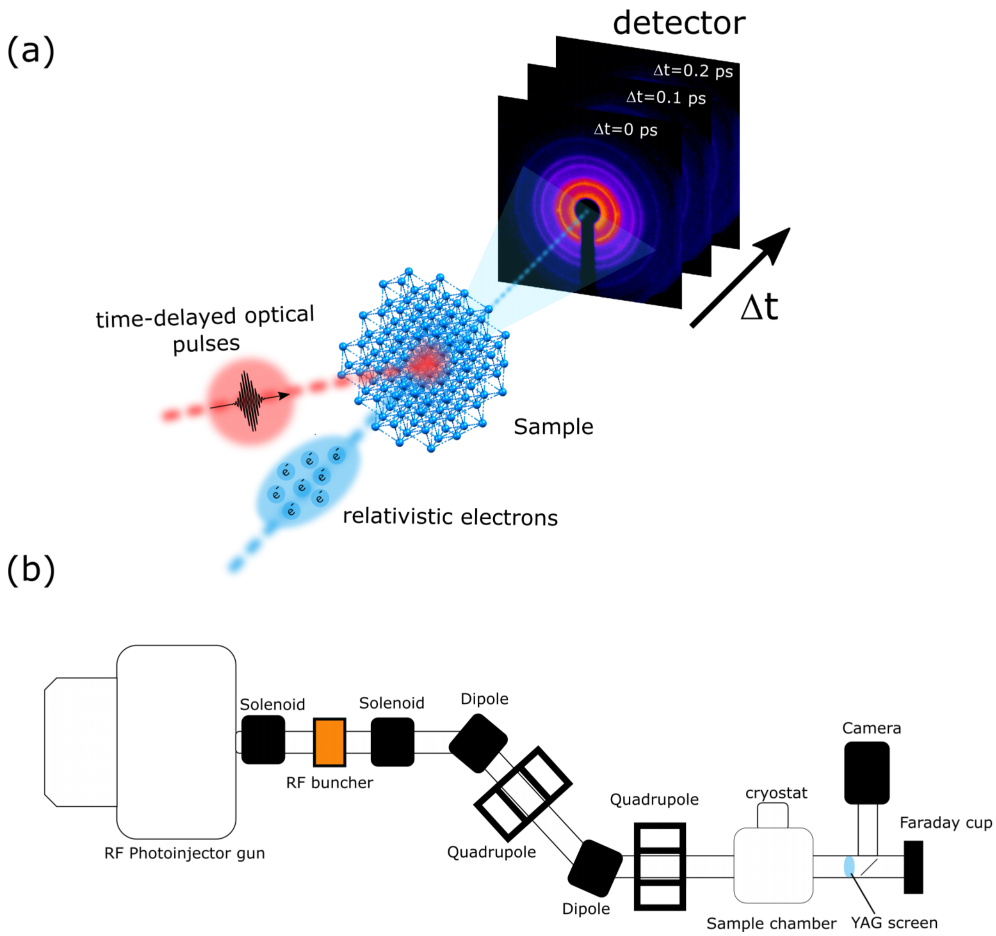

Figure 1. (a) Pump-probe scheme as applied to UED (b) Schematic of HiRES beamline showing key components of MeV-scale electron diffraction setup.

monochromator and collimator for energy selection, and an RF buncher for bunch compression. Other elements such as viewing screens and pinholes are used at various locations along the beamline for alignment purposes. The sample chamber is differential pumped using turbopumps, achieving base pressure of 10^{-7} Torr and houses the sample holder, laser beam steering optics and photodiode for laser/electron diagnostics. A Faraday cup is in place to measure the beam current upstream.

3.1 Sample preparation and characterization

The first step towards UED of materials is acquisition of suitable samples. As mentioned, due to their strong interaction with matter, electrons have high cross-sections for scattering. On one hand this is highly beneficial as weakly scattering specimen, such as gas phase systems, can be studied but, on the other hand, this presents some challenges as solid-state samples (crystalline or amorphous) need to be thin enough to allow transmission of electrons. As a rule of thumb, the thickness needs to be smaller than the mean-free path for elastic scattering, Λ_{el} to avoid multiple scattering effects which can complicate the analysis of the diffraction data.[31] From the pump excitation point of view, thickness must be within the penetration depth of optical absorption so that the sample volume can be homogeneously excited. Typically, samples of thickness in the range of 10-100 nm are needed rendering the sample preparation non-trivial as not all samples can be grown in-situ or their growth controlled. Usually, solid-state systems that are subject of UED studies are first grown as bulk crystals and then thinned down to level required. At LBNL, we use two methods in order to prepare samples, namely mechanical exfoliation and ultramicrotomy. Mechanical exfoliation is a well known technique that has been extensively applied to prepare ultrathin films of 2D materials from monolayers to up to 100 nm thick samples.[32] Here, a

Table 1. Typical operational parameter for HiRES

Parameter	Value
Beam energy	700-900 keV
Repetition rate	single shot $\rightarrow$ 1 MHz
Bunch charge	1-10^8 electrons per pulse
Bunch length	100-1000 fs
Momentum Resolution	0.1 Å^{-1}
Transverse coherence length	$\sim$ 10 nm
Spot size at sample	$50 - 500\mu$m
Momentum transfer range (s)	$\pm 10\text{Å}^{-1}$

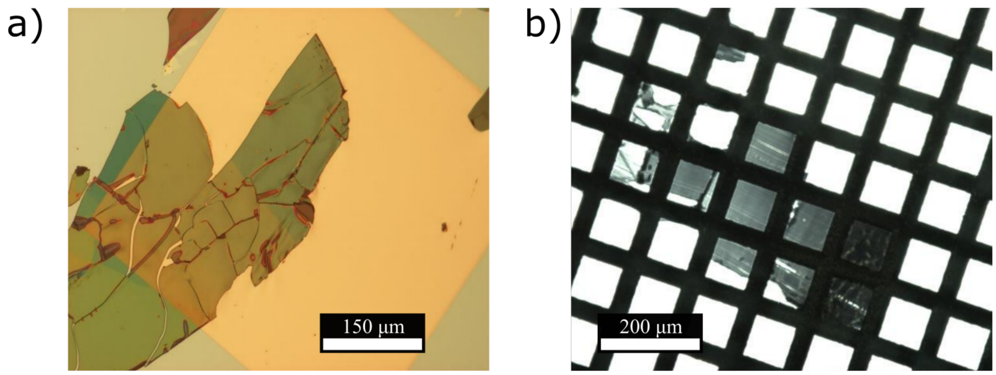

Figure 2. Examples of samples prepared for UED experiments. (a). Bi-2212 film (30 nm) via mechanical exfoliation and dry-transfer method on Si_3N4 and (b). Free-standing TaS_2 (30 nm) on Cu(400) mesh prepared using ultramicrotomy.

scotch tape is used to successively peel layers of van der Waals material until the desired thickness is reached. The thin flake is then transferred first onto a Polydimethylsiloxane (PDMS) stamp and then dry-transferred onto suitable substrate such as silicon nitride, Si_3N_4. Ultramicrotomy, on the other hand, uses a diamond blade to slice the solid specimen down to required thickness with an accuracy of ±5nm. The main advantages of ultramicrotomy are the control of the crystal orientation to cut along and the fact that large area samples ($0.1 - 1000$ mm^2) can be routinely prepared. Figure 2 shows samples of two different materials, Bi-2212 and TaS_2 prepared for UED experiments at HiRES using mechanical exfoliation and ultramicrotomy, respectively. Techniques like atomic force microscopy (AFM) and linear optical spectroscopy provide a means to measure the thickness and equilibrium absorption properties of the system to be studied and are utilized for experiments at HiRES.

3.2 Ultrafast electron diffraction at cryogenic temperatures

Studies of quantum phase transitions encompass a large fraction of condensed matter physics and material science. Many systems that undergo phase transition have at least two stable equilibrium states that are accessible in the temperature range between 10-1000 K. Moreover, most high-T_c superconductors such as the Lanthanum and Bismuth based cuprates have critical temperature, T_c ranging in the 10-100 K, and so their studies require cryogenic capabilities. Having temperature as one of the experimental parameters that can be precisely controlled is, thus, very beneficial and opens up opportunities to study a diverse range of phenomena in materials.

To address this requirement, HiRES is equipped with a closed-cycle liquid helium cryocooler with a vibration isolation interface, which is capable of reaching a stable base temperature of < 10 K. Figure 3 shows the cryostat mounted on the sample chamber in the UED beamline. A specifically designed sample holder is used for ultralow temperature measurements: a copper cassette with slots for samples, alignment pinholes and diagnostic scintillator (YAG) crystal for electron beam as shown Figure 3c. The holder mounts to a multi-axis stage to

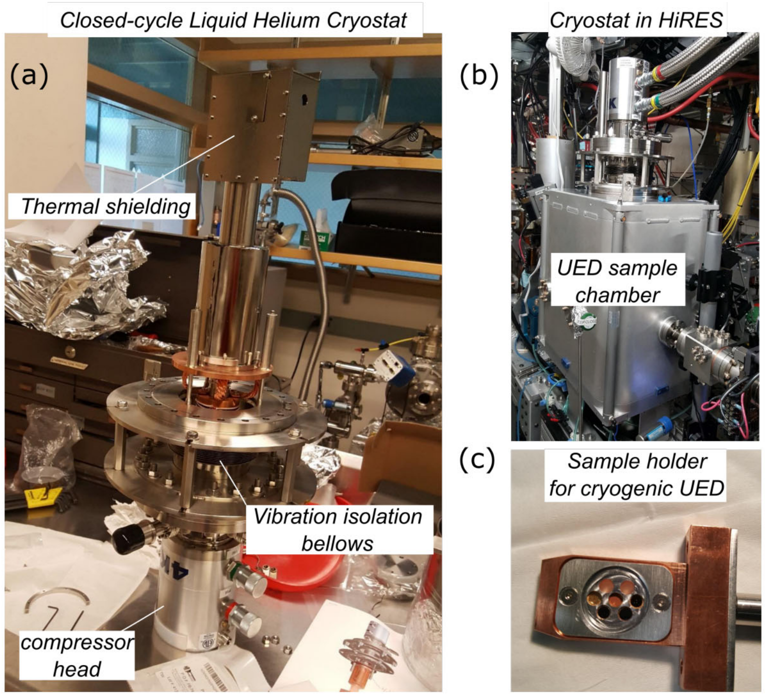

Figure 3. (a) Closed-cycle liquid Helium cryostat for UED studies of materials at cryogenic temperatures. (b) Mounted on UED sample chamber (c). Sample holder for utlralow temperature measurements.

allow sample translation in the Cartesian directions as well as pitch (θ) and yaw (ϕ) rotations. A copper braid connects the cryostat cold-tip and the holder inside a thermally shielded enclosure. Temperature is controlled using a proportional–integral–derivative (PID) temperature controller and read by a silicon-based diode sensor.

Access to ultralow temperatures for UED and a large degree of freedom for sample manipulation ($\theta, \phi = \pm 5°$; x,y,z = $\pm$25 mm) open opportunities for new cases of study involving unconventional superconductors and quantum information systems.

3.3 Pump-probe ultrafast electron diffraction

For measurements of static patterns, electron beam generated by photoemission from CsK_2Sb photocathode using the second harmonic of 1030 nm laser (515 nm) at 250 kHz is directed at the sample using steering electron optics and the diffracted electrons are imaged onto a cerium-doped YAG screen placed about 0.65 m upstream from sample that converts electrons to light flashes, which are then captured by a 16-bit charged-coupled device (CCD) camera with a built-in intensifier. The parameters such as exposure time, gating, integration vs single-shot mode are controlled via the cameras software and are optimized for best signal-to-noise with the shortest integration time. The uniquely high repetition rate of HiRES (up to 1 MHz is possible) means that quick screening of samples can be carried out before pump-probe experiments by collecting diffraction patterns with very high S/N ratio in < 1s.

For pump-probe experiments, a pinhole placed at the sample plane is used to perform spatial overlap between electron and pump laser beams whereas a fast photodiode connected to a fast sampling oscilloscope (20 GB bandwidth) is used to match their arrival times (by scanning the optical delay line) with a resolution of about 100 ps. The electron beam size is controlled using pinholes/electron optics and is typically of the order of 150μm root mean square (RMS) at the sample plane whereas the laser beam size is roughly 800μm RMS, measured using a beam profiler placed outside the chamber at a set distance from sample plane for 1:1 imaging. A standard sample, e.g. bismuth (Bi) film is used to precisely locate time-zero on the delay line. A Matlab software, written in-house and interfaced with CCD camera, is used to control the acquisition of pump-probe data. All elements related to pump-probe measurements can be remotely controlled which makes for a straight-forward and efficient operation.

4. RESULTS

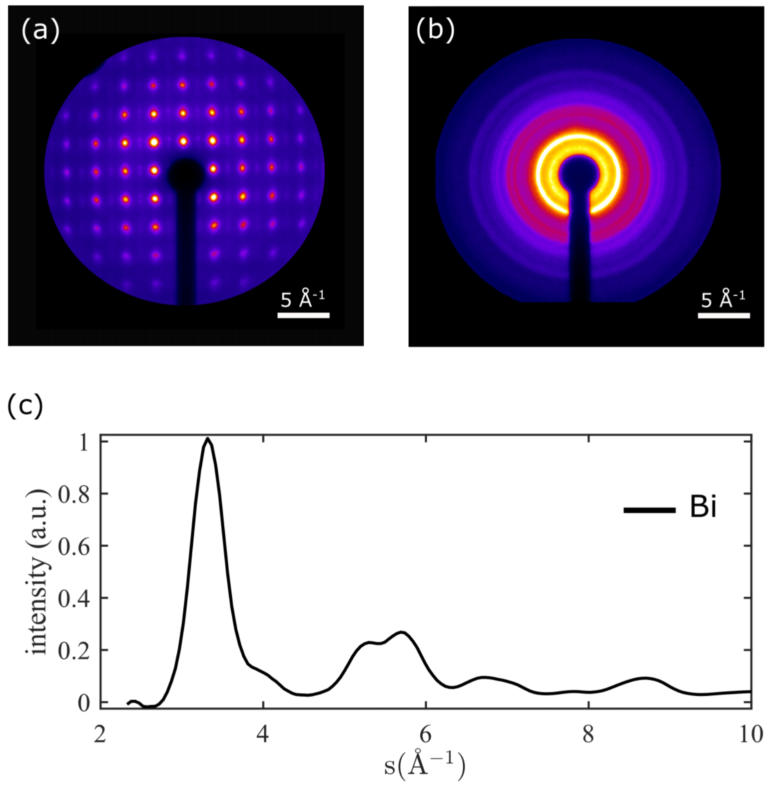

Figure 4. Static Electron diffraction patterns of (a) Bi2212 and (b) Bi(111) measured at HiRES using repetition rate of 250 kHz. (c). Azimuthal integration of diffraction pattern of Bi shown in (b).

Figure 4 shows static diffraction of Bi-2212 (30 nm) and Bi(111) (30 nm) measured at HiRES beamline using relativistic electrons of 0.75 MeV energy. The beam charge for the measurement was 2.5 fC or $\sim$ 15000 electrons per pulse and repetition rate was 250 kHz. The patterns were acquired for about 20 seconds and are an average of 50 frames. The high-quality of the diffraction demonstrates the high S/N ratio obtained due to high brightness and stability of electron source, which is crucial for pump-probe studies. The latter are performed at much lower repetition rates (0.1-10 kHz), a limitation imposed only by the sample recovery times. Top panels of Figure 5 show results of pump-probe experiment on 1T-TaS_2, a benchmark CDW system. Before laser excitation ($\Delta t < 0$), weak superlattice peaks due to CDW order are clearly visible and surround the more intense main Bragg spots, but get strongly suppressed after laser illumination ($\Delta t > 0$) as shown by the middle panel and time-delayed difference pattern. The appearance of the CDW peaks and the high-quality data are evidence for high transverse coherence length of the source (estimated to be $\sim$ 10 nm) and setup performance.

Example of time-resolved measurements at HiRES is given for Bi(111) film of 30 nm thickness excited with 1030 nm laser at 1 kHz. Plots of azimuthally integrated intensities have been stacked together in form of a two-dimensional map. Once again, the high S/N is evident and showcases the quality of time-resolved data that has been measured at HiRES. The time constant, τ_1 recovered from the bi-exponential fit of peak at $s = 5.8\text{Å}^{-1}$ is $\sim$ 350 fs (FWHM), which is in reasonable agreement of 150 fs measured previously reported.[33]

5. CONCLUSIONS AND OUTLOOK

The HiRES beamline at LBNL has been developed for UED studies of materials and isolated quantum systems. With unique capabilities, such as access to a wide range of repetition rates and ultralow temperatures, HiRES is well-equipped for ultrafast studies of conventional and 2D semiconductors both bulk and monolayers, unconventional superconductors and many other interesting systems. Time-resolved UED has been demonstrated

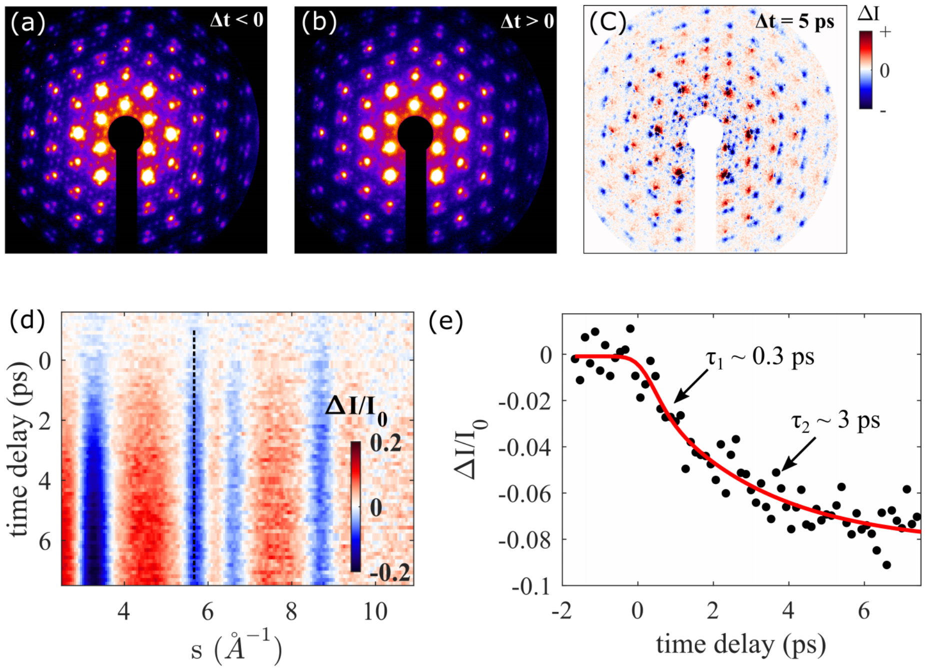

Figure 5. (a) Static diffraction pattern of 1T-TaS_2 (b) 1T-TaS_2 pattern after laser illumination (c) Difference pattern between (a) and (b). (d) Two-dimensional radial map of polycrystalline Bismuth (30 nm) obtained after photoexcitation using 1030 nm laser at 1.5 mJ cm^{-2} (e). Temporal plots of selected peak at $s = 5.8 \text{Å}^{-1}$ denoted by the dotted line and corresponding fit using a bi-exponential function convoluted with instrument response of 750 fs (FWHM).

producing high-quality data and a number of projects have now been completed and projects involving mono-layers and nanofilms are currently being pursued. Further development at HiRES beamline are also underway aimed at electron bunch compression and single-shot experiments, which is expected to further improve the versatility of science carried out at the LBNL UED setup.

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