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VISUALIZING LATTICE DYNAMIC BEHAVIOUR BY ACQUIRING A SINGLE TIME-RESOLVED MeV DIFFRACTION IMAGE*

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Abstract

We explore the possibility of visualizing the lattice dynamic behaviour by acquiring a single time-resolved mega-electron-volt (MeV) ultrafast electron diffraction (UED) image. Conventionally, multiple UED shots with varying time delays are needed to map out the entire dynamic process. The measurement precision is limited by the timing jitter between the pulses of the laser pump and the UED probe. We show that, by converting the longitudinal time of an electron bunch to the transverse position of a Bragg peak on the detector, one can obtain the full lattice dynamic process in a single electron pulse. We propose a novel design of a time resolved UED facility with the capability of capturing a wide range of post-pump dynamic features in a single diffraction image. The work presented here is not only an extension of the ultrashort-pulse pump/long-pulse probe scheme that has been used in transient spectroscopy studies for several decades but also advances the capabilities of MeV UED techniques for future applications with tunable electron probe profile and tunable detecting time range with femtosecond temporal resolution. Furthermore, we present numerical simulations illustrating the capability of acquiring a single time-resolved diffraction image based on the case-by-case studies of lattice dynamic behaviour.

INTRODUCTION

There is a recent surge of interest in studying photoinduced nonequilibrium states of matters. Such studies provide insight into the materials' physical and structural properties out of equilibrium as well as during phase transitions [1]. Searching for new "hidden" phase manipulated by an optical laser pulse becomes increasingly important in condensed matter physics and materials science [2, 3]. The pump laser pulse driving the system out of the equilibrium state initiates the dynamic process. The transient dynamic phenomenon could happen in the timescale of a few to hundreds of ps through electron excitation, thermalization, electron-lattice interaction and phonon-phonon scattering [2-6]. Applying MeV UED with the time-resolving capability to study ultrafast laser-induced long-range structural changes has widely been demonstrated in nanomaterials, 2D materials, bulk crystal samples and even gas phase samples [7-10]. Compared to the conventional 200 keV electron diffraction, the relativistic electron diffraction can be significantly superior in mitigating space-charge effects, packing more electrons in a smaller volume, and penetrating much thicker samples. Compared to X-rays, the MeV UED could access much higher scattering vectors in the momentum space due to the two orders of magnitude shorter de Broglie wavelength of the electrons [7].

In the manuscript, we show how to obtain a single time-resolved diffraction image using a longitudinally shaped electron beam as the probe. Depending on the structure dynamic which leads to the variation of the intensity and/or the position of Bragg diffraction peaks (BDP), we study the correlation of the BDP position and the longitudinal phase space of an electron beam. Based on the correlation, we provide a recipe to convert the arrival time of electrons in a pulse to the transverse position of a BDP on the detector. We study two basic types of pump-probe experiments in structure dynamics, where MeV UED are suitable as the probe, named type-I and type-II cases. To be specific, the type-I experiments refer to the cases in which the material lattice constants have expansion or contraction during the photoexcitation. The changes in crystal lattice constants are often associated with structural phase transition, optical and electronic properties in bulk or nanoparticles through thermal fluctuations [8, 11-22]. Hence the observations of the dynamics of lattice constants are important for many studies in condensed matter physics and materials science. On the other hand, there are photoexcitation processes in solid state materials that the change of the lattice constants may be negligible, but the arrangement of the atoms alters in a unit cell, leading to atomic displacements, symmetry breaking and other types of lattice distortion. As result, the reflections have intensity variations due to the structure factor change as a function of time [4, 9, 20, 21]. This is the type-II case in our study. Therefore, our measurements focus on the BDP positions in the reciprocal space due to lattice constant change after the pump in the type-I cases, while we concentrate on the BDP intensity measurements in the type-II cases. Although lattice dynamics change many BDPs, we only focus on the variation of the intensity and/or the position of one BDP, which is chosen for probing the photoexcitation dynamics. Our goal is to utilize the post-pump dynamic features to create a single-shot solution, which is applicable to all cases including type-I, type-II, and hybrid where both the intensity and position of a BDP change.

The electron bunch is constituted by a sequence of beamlets continuing in time typically with a Gaussian envelope. The timescale and the required temporal resolution of

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structure dynamics determine the bunch length and the beamlet duration, respectively. Each beamlet represents a sample point in time. The diffraction signal from each beamlet-and-sample interaction forms a diffraction pattern. Due to the limited speed of the detector, the recorded diffraction image is the superposition of diffraction patterns from all beamlets with different time delays usually having a sub-ps timescale. This problem can be overcome by mapping the time delay of all beamlets to the different transverse positions of the BDP via longitudinally shaping the electron beam and introducing a time-dependent transverse kick after the sample diffraction. After the pump excitation, the intensity and position of a BDP usually vary with time, ranging from sub- to tens- ps. Both the desired femtosecond resolution in a sub-ps timescale and the tunability for various timescales represent significant challenges. Thanks to our novel design, those needs can be satisfied by using two independently adjustable electromagnetic (EM) chicanes [22]. To minimize the dose of electrons, the first chicane, positioned upstream of the sample, is optimized to match the electron bunch length to the lattice dynamic timescale. All the electrons diffracted by the sample carry the dynamic information. The second chicane, located downstream of the sample, can be independently adjusted to make the bunch sufficiently long, therefore only requiring a moderate deflecting voltage. The timescale match is especially important for the ultrafast sub-ps phenomenon, which requires a ten-fs temporal resolution. Furthermore, a novel scheme of bunching electrons at the period of hundreds femtosecond to a few ps allows the experimental study of the mechanism of radiation damages in such short timescales and enables UED as the probe for beam-sensitive soft materials.

RESULTS

Streaking UED

Mapping the time of an electron bunch to the transverse position of a BDP on the detector is common for both type-I and type-II cases. Comparing type-I and type-II post-pump dynamic features, the BDP intensity variation in the type-II case is simple and straightforward. An adequate angular resolution is needed for resolving the BDP position variation in the type-I case. Therefore, we start the design of a single time-resolved UED probe in the type-I case (BDP position vs. delay time) and it should be applicable to the type-II case.

The BDP position on the detector moves in responding to the lattice constant variation with time after the pump excitation, obeying Bragg's law $2d \sin\theta = n\lambda$, where θ is the incident angle, d is the crystal interplanar distance proportional to the lattice constant, λ is the de Broglie wavelength, n is the reflection order. The lattice constant $d(t)$ and the BDP position $y(t)$ can be described by Eq. (1):

$$y(t) = L_{S2D} \tan[2\theta(t)] \approx L_{S2D} \frac{n\lambda}{d(t)} . \quad (1)$$

Here, $t = 0$ is the time when the pump laser pulse arrives at the sample and L_{S2D} is the distance from the sample to the detector. We assume the duration of the ultrafast pump laser pulse is negligible compared to the total duration of the electron beam. The y coordinate on the detector starts from the center of the diffraction image and points to the direction of the BDP, which is chosen for probing the post-pump dynamics.

Experiments with a single time resolved UED image will require the total length of the electron beam T comparable to the timescale of the post-pump dynamic (e.g. 15 ps). The trajectory of the BDP position on the detector is determined by the post-pump dynamic *via* $d(t)$. The reference of the BDP intensity profile without the pump is proportional to the current distribution $I_c(t)$ of the incident electron beam. Here, $I_c(t_i)$ is the charge density of the i^{th} beamlet, $i = 1, 2, \ldots, N$. $N = T/\Delta t$ is the total number of beamlets, where Δt is the beamlet duration. Any change of the BDP position and intensity profile, relative to the reference, contain the information of the lattice evolution excited by the pump laser pulse. This information can be used to map out the entire dynamic process of the system.

Thus, acquiring a single time-resolved UED image to capture time-resolved lattice behaviour becomes possible. However, there are some technical challenges that must be overcome before the single-shot time-resolved UED probe can deliver the sub-picosecond to femtosecond resolution. The lattice constant variation $\Delta d/d$ (typically in the range from 10^{-3} to 10^{-2} for many metal and oxide materials) determines the range of BDP motion on the detector:

$$\frac{\Delta y}{y} = -\frac{\Delta d}{d} , \quad (2)$$

which is usually from sub-pixel to a few pixels, depending on the detector setup. To resolve each individual beamlet in the timescale of ten-femtosecond to sub-picosecond, N needs to be as big as few hundred (e.g. $N = T/\Delta t = 15\ \text{ps}/0.05\ \text{ps} = 300$). Since at least 3 pixels are necessary to resolve each point on the detector [23, 24], the range of BDP motion is required to be larger than $3N$ pixels.

A deflecting cavity is routinely used to map the longitudinal profile of an electron bunch to the transverse profile of an image on the detector [15-17] (Fig. 1). To minimize errors introduced by the deflecting cavity (e.g. phase jitter), we prefer to map the time profile of the bunch to the x-direction, orthogonal to the direction of BDP motion, by rotating the sample. The $x(t)$ projection described by Eq. (3) carries the time information with a precision better than ten femtosecond [14-16]:

$$x(t) = L_{C2D} \tan[\theta_x(t)] , \quad (3)$$

where

$$\theta_x(t) \approx \frac{eV_\perp}{pc} \omega_{RF} t \quad (4)$$

is the deflection angle; L_{C2D} is the distance from the deflecting cavity to the detector; $V_\perp$ is the deflecting voltage; ω_{RF} is the RF angular frequency; p is the longitudinal momentum of the electron beam.

The required time resolution is achieved by making the motion range of the electron bunch $\sqrt{\Delta x^2 + \Delta y^2} \geq 3N$ pixel. Here Δy and Δx represent the BDP motion caused by the change of lattice constant excited by the pump pulse and the RF

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deflection, respectively. The maximum deflection $\Delta x = x(T) - x(0) = L_{C2D}\tan[\theta_x(T)]$ is tunable via varying the deflecting voltage $V_\perp$ while Δy is relatively small. To achieve $\Delta x \gtrsim 3N$ pixel ($\Delta y \ll \Delta x$), the required voltage estimated from Eq. (4) is usually in the range of a few to tens kilovolt while $\theta_x(T)$ is about a few to ten milliradians.

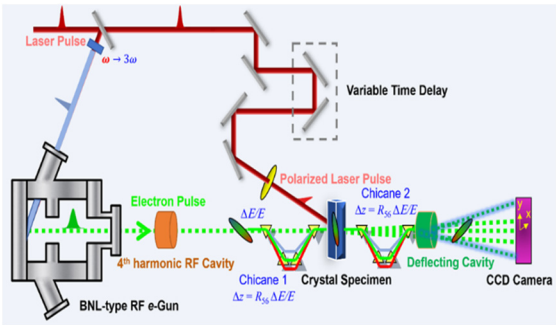

Figure 1: Schematic layout of the single time-resolved pump-probe experimental setup. The 4th harmonic RF cavity controls the energy chirp of the electron beam before the sample; the deflecting cavity performs time-to-position mapping after the sample. To achieve the best possible time resolution, two EM chicanes placed before (Chicane 1) and after (Chicane 2) the sample are responsible for real-time tuning and matching the electron bunch length to the lattice dynamic timescale.

Numerical Studies Case-by-Case: Hybrid

The numerical example of a hybrid case (e.g. both BDP position and intensity vary as a function of time) is chosen. The simulations are done with the beam charge of 6 pC and the flat-top longitudinal profile. In addition to the BDP intensity variation after the pump excitation, the lattice constant varying with time exponentially from 5 to 5.25 Å in 15 ps. With the optical pump, the vertical and horizontal positions of the BDP in the diffraction image contain the information of the lattice dynamic motion and the delay time relative to the arrival of the pump pulse, respectively. Figures 2 (a) and (b) show the time-resolved diffraction image on a detector with a pixel size of 5 μm and 1 μm, respectively. The results of peak finding in y at every time slice are shown as blue (1 μm) and black (5 μm) curves in Fig. 2 (c). The red curve represents the time dependent BDP position resulted from the ideal model of lattice dynamics after the pump laser pulse. Similarly, the results of integrating the image intensity along y at every time slice are shown as blue (1 μm pixel) and black (5 μm pixel) curves in Fig. 2d. The red curve represents the time dependent BDP intensity resulted from the ideal model. The simulation result confirms the detector pixel size must be ≤ 3 μm for the required resolution.

For both type-I and type-II pump-probe experiments, the slope of the energy chirp introduced by the BNL-type photocathode RF gun is constant and independent of the electron bunch length at the gun phase ranging from 6° to 25°. An X-band (4th harmonic) RF cavity will be used to remove the chirp. Both the linear chirp of the electron beam energy with respect to the gun phase and the linear amplitude-phase range of the 4th harmonic cavity limit the maximum timescale of the pump-probe experiment to ≤ 15 ps [25]. Controllable energy chirp in combination with a magnetic delay line can extend the timescale coverage somewhat larger. Without changing the current photocathode gun, we still need a few shots of 15 ps long electron beams with various delays to achieve longer timescales. However, it is still a significant improvement in comparison with conventional pump-probe experiments requiring hundreds of UED shots.

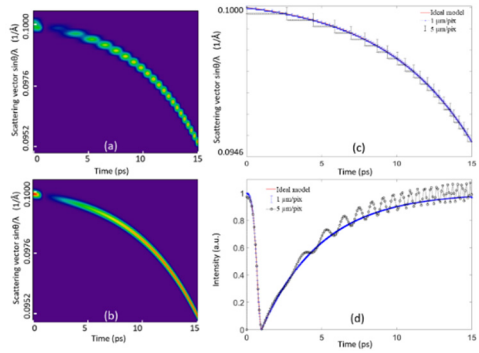

Figure 2: Combined post-pump dynamic of lattice constant variation and BDP intensity variation: time-dependent diffraction images after the pump on the detector with pixel size of 5 μm (a) and 1 μm (b) representing the BDP intensity and position; results of peak finding at every time slice in x (c); results of intensity integration at every time slice in x (d).

CONCLUSION

The temporal resolution of the pump-probe UED experiment continuously improved in the past several years. The main advantage of our novel time-resolved single-shot UED is to maximize the signals for visualizing the lattice dynamical behavior and minimize the sample damage induced by the pump laser, this will help in having all the data taken for the entire dynamic process with much fewer laser pumping than multiple-shot time-resolved techniques. We lay out a comprehensive design for laser-pump UED-probe experiment toward single-shot time-resolved electron diffraction and microscopy. The hybrid, type-I and type-II experiments can share the same setup. To achieve the best possible temporal resolution, we implement a real-time tuning scheme for the best matching between the electron bunch length and the lattice dynamic timescale, ranging from sub-picosecond to tens picoseconds. Furthermore, a state-of-art superconducting photocathode RF gun which can extend the electron bunch length to hundreds picosecond is in the process of being built [26]. As result, the entire timescales of structural dynamics can be covered with a single shot. The resolution of post-pump dynamics can be further improved using a real-time longitudinal beam profile monitored via the temporal profile of the driving laser, similar to phase retrieval with the pre-knowledge of the beam current profile [26, 27]. However, to fully benefit from this novel design, several technical challenges, e.g. synchronizing different RF cavities with the required temporal accuracy, must be fully realized in experiments.

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