

Streak-camera reflection high-energy electron diffraction for dynamics of surface crystallography



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ABSTRACT

A new technique for ultrafast dynamics of surface crystallography was developed by combining reflection high-energy electron diffraction with the electron deflectors of a streak camera system. A one-dimensional distribution of electrons scattered by a crystal surface is selected by a linear slit on a screen, and then the electrons are quickly deflected by the sweep electrodes behind the slit. Thus, a temporal evolution of the one-dimensional diffraction pattern can be displayed as a streak image on a screen. This is a unique method of time-resolved electron diffraction, as a pulsed electron beam is not required to obtain a temporal evolution. The temporal evolution of the diffraction pattern can be projected on a screen from single-shot measurements. The technique was tested on an Si(111)- 7×7 surface, and the dynamics of the surface structure were successively obtained from changes in spot intensities. Although the present time resolution was limited by the present pumping laser ~ 5 ns, the nominal resolution of the streak system is expected to be ~ 100 ps.

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1. Introduction

In order to investigate the dynamics of a surface, it is crucial to understand the temporal changes in both electronic and atomic structures. Progress in femtosecond laser technology has provided optical pump probe techniques for studying ultrafast phenomena induced by light emission [1]. For example, spectroscopic techniques and photoemission with ultrafast resolution are available for observing the dynamics of electronic systems [1–6]. Although the atomic movement might be speculated from studies of the ultrafast spectroscopy, it is straightforward to observe structural dynamics using time-resolved diffraction. The time-resolved [7–12] techniques have been extensively developed over the past 15 years, such as time-resolved X-ray diffractions [7–13] and time-resolved electron diffractions [7,8,14–21]. Recent progress in time-resolved diffraction techniques has allowed the observation of lattice dynamics, even on a femto-seconds timescale [11–21]. In general, the short pulse probe, i.e. X-rays for XRD [10, 12] or electrons for electron diffraction (ED) [22–24], is the key to establishing a high temporal resolution for the pump and probe techniques, where the temporal evolution for the range from fs to ns is surveyed by an optical delay line. Sophisticated and large-scale apparatus have been developed to provide the ultra-short pulses for the time-resolved XRD or ED.

However, the ultra-short pulses used in such methods require a large-scale apparatus, i.e. rf electron gun [8] or X-ray free electron

laser [9] to perform time-resolved diffraction. The temporal resolution and the range required for the time-resolved diffraction vary depending on the targeting phenomena. A time resolution of < 1 ps is essential for capturing ultrafast atomic motion and phonon oscillation. Picosecond resolution is usually sufficient to study structural changes in thermal processes. Furthermore, a process involving atomic diffusion on the surface would show a much slower evolution of the diffraction patterns. In order to investigate such slow processes, the technique that can access a wide range of time scales from ns to ms with a moderated resolution would be extremely useful, since the pump-probe techniques are suitable to probe the ultrafast range from fs to ns.

A streak camera, which can capture rapid temporal evolution of photon signals, is an alternative to the pump-probe technique with a short pulse probe [25,26]. An incident photon signal is converted into electrons by a photocathode in the streak camera, and then the electrons are accelerated and quickly deflected by the sweeping field. Thus, the time-evolution of the incident photon intensity is displayed on a fluorescent screen as a streak image. In principle, the time evolution can be obtained from single-shot measurements, whereas multi-shot measurements with varying delay times are required for the pump probe technique with a pulse probe. The duration of the streak is easily changed by altering the sweep speed of the electrons, which can be electronically controlled, typically from milliseconds to hundreds of picoseconds. A streak camera with a temporal resolution of a few hundred femtoseconds is commercially available, although the signal intensity becomes weak as the scan speed increases. It is advantageous that a streak camera can cover a wide range of time scales and in principle does not require a short pulse probe.

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The temporal resolution and the range required for these measurements vary depending on the targeting phenomena. A time resolution of <1 ps is essential for capturing fast atomic motion and phonon oscillation. However, picosecond resolution is usually sufficient to study structural changes in thermal processes. Furthermore, a process involving atomic diffusion on the surface would show much slower evolution of the diffraction patterns. An easy-to-use time-resolved diffraction technique with access to a wide range of time scales would be extremely useful.

Here, we have developed a novel time-resolved electron diffraction technique in which reflection high-energy electron diffraction (RHEED) and a streak camera are combined into a unified apparatus. Although the streak camera has been used to measure the duration of electron pulses [16,27] and to improve the temporal resolution of the pump-probe electron diffraction [28], the present study is the first attempt to combine it with the RHEED for dynamics of the surface crystallography.

Although streak cameras were used to measure the duration of electron pulses [16], they have not yet been used to measure the time evolution of a 1D diffraction pattern. We prepared streak electrodes in a UHV chamber of RHEED. High-energy electrons scattered by the sample surface pass directly into the streak electrodes, and they are then swept to show the temporal evolution on a screen. There is no photo-cathode in this apparatus. We call the method streak-camera RHEED (SC-RHEED). It does not require a short pulse probe for high temporal resolution, so the equipment is very compact in comparison with other time resolved XRD or ED method that employs a short pulse probe. We demonstrate the new technique on an Si(111) surface with a pulse laser (~ 5 ns) to pump a structural transition. Although the nominal temporal resolution of the streak system was estimated to be ~ 100 ps, the present resolution was limited by the pulse width of the laser. The resolution can be improved by using a shorter, sub-picosecond pumping pulse laser. Although the present method cannot provide the ultrafast resolution, it can cover a wide temporal range from ns to ms to investigate the evolution of the diffraction patterns. The present SC-RHEED is a promising tool for studying structural dynamics on crystal surfaces.

2. Principle and design

A schematic drawing of the SC-RHEED apparatus is shown in Fig. 1(a). As in a standard RHEED system, a high-energy electron beam, typically 10 keV, is incident on a crystal surface with a grazing incidence. Electrons scattered by the surface form a diffraction pattern on a front screen, which is located at the entrance of the streak unit.

The front screen, as schematically shown in Fig. 1(b), has a linear slit to admit electrons into the streak unit so that a distribution of electrons, i.e. a one-dimensional (1D) diffraction pattern, can be introduced into the unit. Then, the electrons are quickly swept perpendicularly to the slit by the streak deflectors. When the sweep is synchronized with the a structural pumping, time evolution of the 1D diffraction pattern is displayed on an MCP screen as streaks, as shown schematically in Fig. 1(c). A laser with nano- to femto-second pulse duration is suitable to pump the structural phase transitions.

In order to optimize the design of the electrodes of the streak unit, we performed a numerical simulation of electron trajectories using the simulation software SIMION [29]. The basic design of the streak unit is shown in Fig. 2. As shown in the top view, the unit accepts electrons from a sample with an azimuthal angle range of $\sim 27^\circ$. The front screen, slit, lenses and streak deflectors are sections of concentric cylinders whose center is at the sample, so that the azimuthal angle of the electron is preserved from the sample to the MCP screen. As seen in the side view, electrons scattered by the sample form a diffraction pattern on the front screen, and its 1D section is selected by the linear slit. The distance from the sample point to the slit was 30 mm, and the slit gap was 0.2 mm. Electrons passing through the slit are deflected by the lenses to enter the gap between the streak deflectors. In order to drive the streak deflectors, we used an electronic power supply from a commercial streak camera (C7700, Hamamatsu Photonics K. K.). The power supply provided sweep voltages from -500 V to $+800$ V and from $+500$ V to -800 V for the lower and upper deflectors, respectively, which were intended to deflect electrons accelerated by ~ 8 keV in the original streak tube. RHEED electrons with a kinetic energy of 10 keV are deflected by the sweep voltages in the present unit. We can electronically select the duration of a full sweep from ~ 2 ms to ~ 1 ns at a maximum repetition of 1 kHz by adjusting the power supply. An MCP screen (3 in. in diameter) was placed 155 mm from the sample. The $\sim 27^\circ$ acceptance angle of the screen is typical of standard RHEED optics. All components and their arrangement were designed on the basis of simulated electron trajectories.

As seen in the side view, the MCP screen was surrounded by a shield to prevent electrons from entering the screen unless they were swept. Once the electrons are swept downward from the upper initial position to the final position of the sweep, they must return to the initial position for the next sweep. Since the electrons can also enter the screen during the return sweep, we installed blanking gate electrodes to the electron gun so that the electron beam would not enter the sample or the unit during the return sweep.

In the simulation, we treated the electric field as a static one. However, we have to consider dynamics of the electric field in the streak

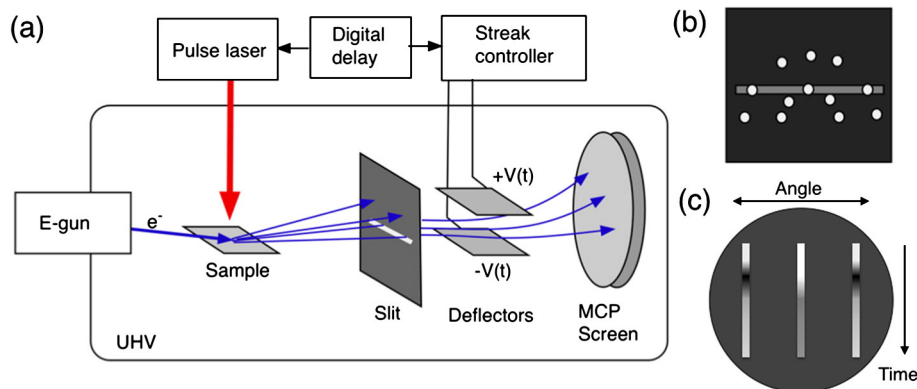


Fig. 1. Schematic diagrams of streak-camera RHEED: (a) system diagram, (b) diffraction pattern on the front screen with the slit, and (c) streak image on the MCP screen, i.e. the time evolution of the diffraction pattern.

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