Ultrafast x-ray diffraction using a streak-camera detector in averaging mode

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We demonstrate an apparatus for measuring time-dependent x-ray diffraction. X-ray pulses from a synchrotron are diffracted by a pair of Si(111) crystals and detected with an x-ray streak camera that has single-shot resolution of better than 1 ps. The streak camera is driven by a photoconductive switch, which is triggered by 100-fs laser pulses at a repetition rate of 1 kHz. The laser and the streak camera are synchronized with the synchrotron pulses. In the averaging mode, trigger jitter results in 2-ps temporal resolution. We measured the duration of 5-keV pulses from the Advanced Light Source synchrotron to be 70 ps. © 1997 Optical Society of America

Laser sources currently produce ultrashort (<1-ps) pulses at infrared, visible, and ultraviolet wavelengths that can be used for study of the dynamics of valence electrons in atoms, molecules, and solids. However, for direct probing of structural properties of molecules and solids it is advantageous to use x-ray radiation, and ultrafast x-ray techniques and detectors have only recently become available. For example, conformational changes were recently studied in the nanosecond time domain at the European Synchrotron Radiation Facility, using Laue diffraction from a rapidly evolving myoglobin sample.[1] The time resolution in that experiment was limited by the duration of a single pulse of x-rays from the storage ring. With improved resolution in the range of 1 ps, x-ray-diffraction techniques could be applied to the study of ultrafast processes such as laser-induced phase transitions and transient excited modes in solids.

We have established an x-ray experimental facility at the Advanced Light Source (ALS) synchrotron at Lawrence Berkeley National Laboratory, which will permit the study of ultrafast structural dynamics. These dynamics are initiated by 100-fs laser pulses from a $Ti:Al_2O_3$ laser system, which is synchronized to electron bunches in the storage ring at the laser repetition rate of 1 kHz. The duration of x-ray pulses from the ring varies with the mode of operation but is typically 30–80 ps. An x-ray streak camera,[2] which is capable of detecting single photons in a single-shot mode and with subpicosecond temporal resolution, is triggered by the laser pulses. The timing jitter of the camera relative to the laser is ~2 ps. This diagnostic will enable us to study x-ray-diffraction signals that are changing rapidly in both time and momentum space. Such a changing signal is expected, for example, from the disordering of a crystalline lattice following ultrafast deposition of laser energy.

Our apparatus is shown in Fig. 1. The ALS bending magnet (Beamline 10.3.2) emits a broad spectrum of x-ray radiation, extending to a useful photon energy of ~12 keV, when the electron energy in the ring is 1.9 GeV. Experiments were performed with the ALS operating in a double-bunch mode, for which the average beam current is 40 mA and the pulse repetition rate is 3 MHz. The divergence of the x-ray beam is 0.4 mrad, as determined by a round aperture at the end station that contained the experiment, which was 30 m from the ring. Broadband radiation is monochromatized by a parallel pair of Si(111) crystals separated by 1 m. The crystals diffract in the vertical plane, corresponding to *s* polarization. The Bragg angle was 22.5°, corresponding to the reflection of 5-keV x-rays. We maximize the signal seen by the streak camera by use of a first crystal that is sagitally

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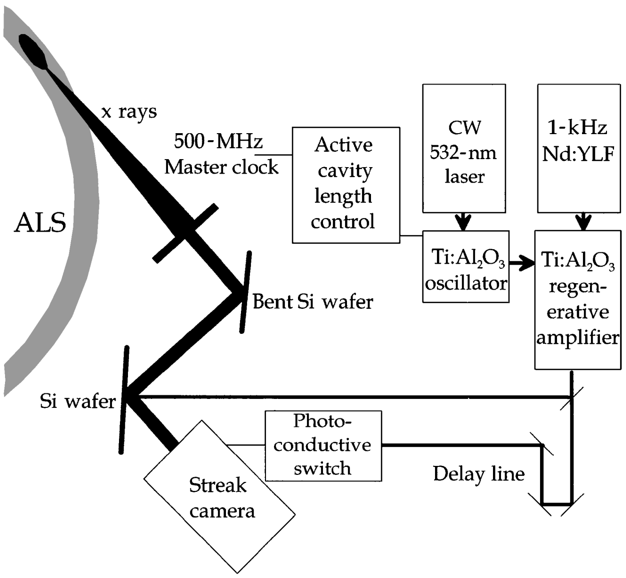

Fig. 1. Experimental setup showing the x-ray radiation from the synchrotron, two Si crystals, laser and triggering systems, and the streak-camera detector.

curved, which results in a line focus of the x-ray radiation on the entrance slit of the streak camera. The camera is positioned approximately 10 cm from the second crystal. We bent the first crystal, a 300-μm-thick Si wafer, by gluing it onto a cylindrically machined Al mandrel. The slope error of the surface of the wafer in the diffraction plane was ~1 mrad, which is ~10 times the width of the rocking curve for the designated reflection. A horizontal focal width of 0.4 mm was measured on thermally sensitive paper placed on top of the second crystal. We measure 1500 diffracted photons per synchrotron pulse, using an x-ray-sensitive Si photodiode in place of the streak camera. We expect to be able to obtain flux levels approximately one order of magnitude higher flux levels than reported in this study by using a bent crystal with less slope error.

The laser system consists of a Ti:Al$_2$O$_3$ oscillator pumped by a frequency-doubled Nd laser (Millenia, Spectra Physics) and a Ti:Al$_2$O$_3$ regenerative amplifier system (Spitfire/Merlin, Positive Light) operating at a repetition rate of 1 kHz. The pulse duration is ~100 fs, and the pulse energy is ~1 mJ. We synchronize the laser pulses with the synchrotron pulses by locking the laser-oscillator repetition frequency (set by the oscillator cavity length) to the master clock of the ALS.[3] From the error signal observed in the stabilization electronics, we estimate the jitter between the laser and the synchrotron pulses to be ~5 ps. This residual jitter is unimportant for our experiments, since the ALS pulse has a much longer pulse duration.

Using a commercial x-ray streak camera (Kentech low-magnification model), we were able to detect single synchrotron pulses, as shown in Fig. 2. However, the signal was relatively weak. This results from (1) the relatively low flux from a simple bending magnet source and (2) the low quantum efficiency (~1%) of the 100-nm-thick CsI photocathode. Low quantum efficiency is a well-known limitation in the use of streak-camera detectors that must be considered along with their unique high-speed response. The resulting measurement of the ALS pulse duration is ~80 ps.

A higher signal-to-noise ratio can be achieved in two ways. First, for single-shot experiments, the higher flux levels available from a modern synchrotron-insertion device such as a wiggler or undulator will increase the signal by several orders of magnitude and permit single-shot detection of diffracted x-ray signals. A second approach (demonstrated in our research) involves averaging over multiple pulses. However, if this approach is used, the temporal jitter associated with triggering of the streak camera must be less than the required time resolution. One can achieve this by triggering the sweep voltage to the deflection plates of the streak camera with a laser-triggered photoconductive switch.[4] We therefore employed a second x-ray streak camera,[2] which is capable of operating at a high (1-kHz) repetition rate (for increased averaging), high time resolution (880 fs), and low jitter triggering (when a photoconductive switch is used). Recently this camera was shown through the use of an improved sweep plate design and a unique high-voltage pulser to have a resolution of 540 fs.[5]

Jitter in the streak-camera timing with respect to the laser pulse was measured with a test setup. We used ~20 μJ of 800-nm light from the Ti:Al$_2$O$_3$ laser to trigger a GaAs photoconductive switch, which drove the high-voltage pulse to the sweep plates of the streak camera. We obtained UV light with a wavelength of 266 nm by frequency mixing the 800-nm light in two KDP crystals. The UV light was split into two pulses separated by 20 ps by use of a fused-silica window of known thickness, which was inserted halfway into the beam. This UV double pulse was then incident upon the photocathode and was used for calibrating the time response and the relative jitter of the streak camera. As shown in Fig. 3, the resulting effective temporal resolution of the streak-camera system was 2 ps in this averaging mode (here ~5000 pulses were summed).

As a test of our ability to observe time-resolved events in the x-ray spectral region, we measured the duration of the diffracted ALS pulses. The result of the measurement is shown in Fig. 4. We obtained the data by averaging over 2000 pulses. The observed

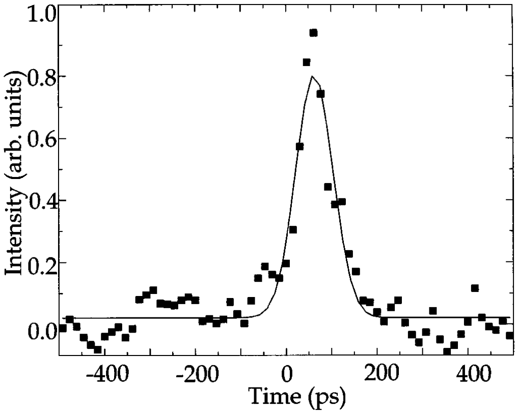

Fig. 2. Temporal profile of a single ALS pulse, measured with the Kentech streak camera.

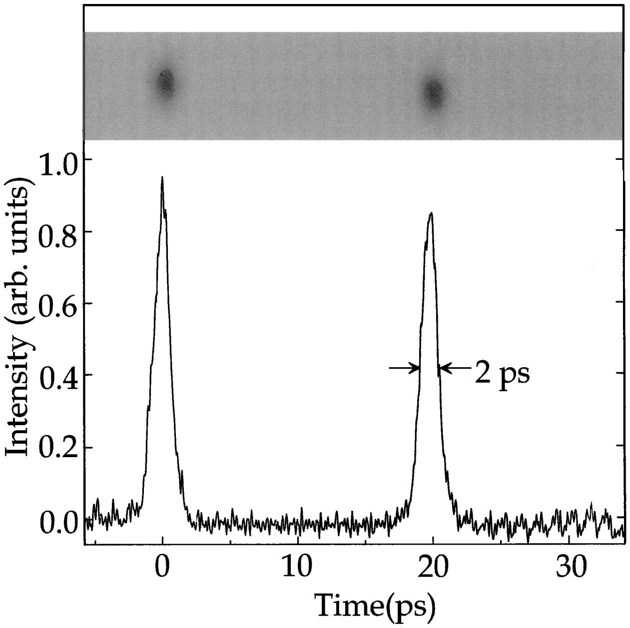

Fig. 3. Temporal response of the ultrafast, photoconductive-switch-triggered streak camera was measured to be 2 ps when 5000 pulses were averaged.

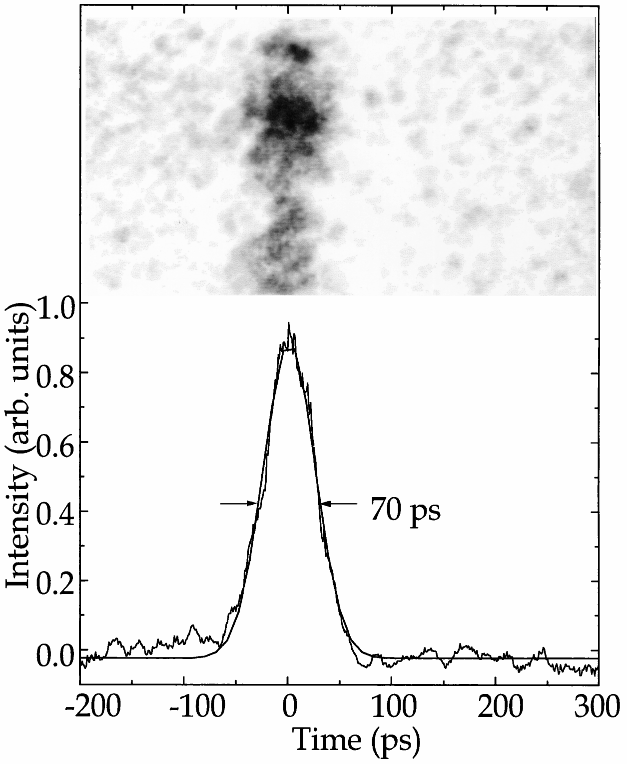

Fig. 4. Pulse duration of the ALS was measured to be 70 ps with a streak camera operating in an averaging mode. Both the camera output and a lineout of these data are shown.

pulse duration of 70 ps is in good agreement with previous measurements of the ALS pulse duration.[6] In future experiments we expect to be able to observe substructure within the 70-ps time window defined by the ALS x-ray pulse, following illumination and heating of the sample (the second Si crystal) by the ultrafast laser pulse.

In conclusion, we have demonstrated an x-ray streak-camera detector system that is synchronized to both an ultrafast laser system and the x-ray pulses from a synchrotron. The camera has temporal resolution of 2 ps when operating in an averaging mode for events that are triggered by ultrafast laser pulses. Our setup will allow us to pursue laser-pump/x-ray-probe experiments for monitoring of structural changes in materials with ultrafast time resolution.

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