Visualization and control of atomic motion

A tunable undulator source to generate femtosecond x-rays is now in operation at the SLS storage ring. The 2.5–1 Å output radiation is driven by an initial 50 fs laser pulse employing the laser-electron slicing technique [1]. It provides inherently synchronized femtosecond laser 'pump' and x-ray 'probe' pulses to enable time-resolved absorption and diffraction experiments. A maximum sliced x-ray flux of 2 x 10^5 photons per second and 0.1% bandwidth was measured at 5 keV. Despite this rather low photon flux it is demonstrated that extended pump-probe scans can be performed due to an outstanding spatial and temporal stability of the electron, x-ray and laser pulses. This excellent stability has become possible by taking advantage of the highly stable electron beam operated in top-up mode and controlled by a fast orbit feedback system in combination with state of the art laser systems.

Whereas ultrashort laser pulses are commonly characterized using intensity autocorrelation techniques, a standard method to measure the duration of femtosecond hard x-ray pulses has yet to be developed. Here the temporal characteristics of the x-ray pulses are determined studying high-amplitude phonon dynamics of photoexcited bismuth. Optical excitation of the near-surface region of the crystal by an intense femtosecond laser pulse results in a fast initial displacement and consecutive oscillation of the atoms around their new equilibrium position. This coherent large amplitude motion of the atoms along the body diagonal of the rhombohedral unit cell (see Fig. 1(a)) can be observed by measuring the diffracted intensity from certain Bragg reflections as a function of time [2]. Grazing incidence diffraction (~0.4 - 2º) is used to vary the probe depth in the crystal. Close to the sample, a single Kirkpatrick-Baez mirror focuses the beam vertically to a size of 10 μm and a single multilayer monochromator restricts the bandwidth of the incoming pink undulator beam. In this configuration the incident flux is typically ~200 photons per pulse in a 1.2% bandwidth. The pump laser is incident on the sample at a grazing angle of 14º limiting the time smearing effect of the noncollinear geometry to approximatly 100 fs.

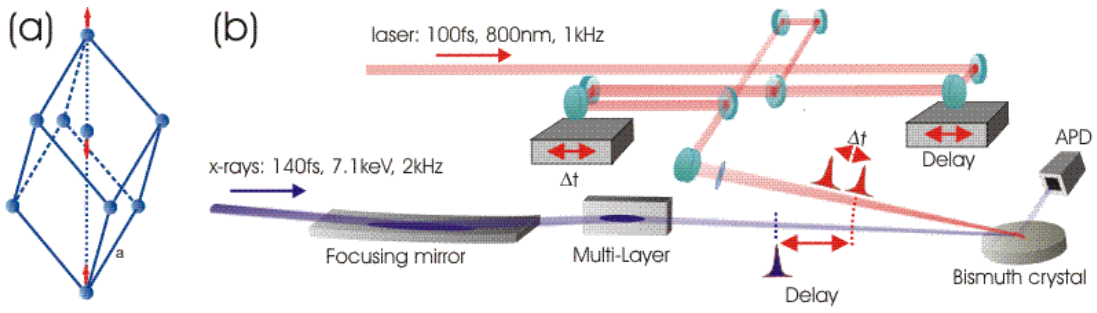

Figure 1: (a) *Bi unit cell structure. The A_{1g} phonon motion of the atoms is indicated by the red arrows.* ***(b)*** *Setup of the time-resolved diffraction experiments at the μXAS-beamline at SLS.*

The response following excitation by a single laser pulse is shown in Fig. 2(a) for an absorbed fluence of 1.12 mJ/cm2 displaying an effective time resolution of 195 ± 25 fs FWHM. Deconvolution with the duration of the pump pulse and the noncollinear geometry yields an effective x-ray pulse length of 140 ± 30 fs FWHM. Timing drifts measured over several days are below 30 fs rms.

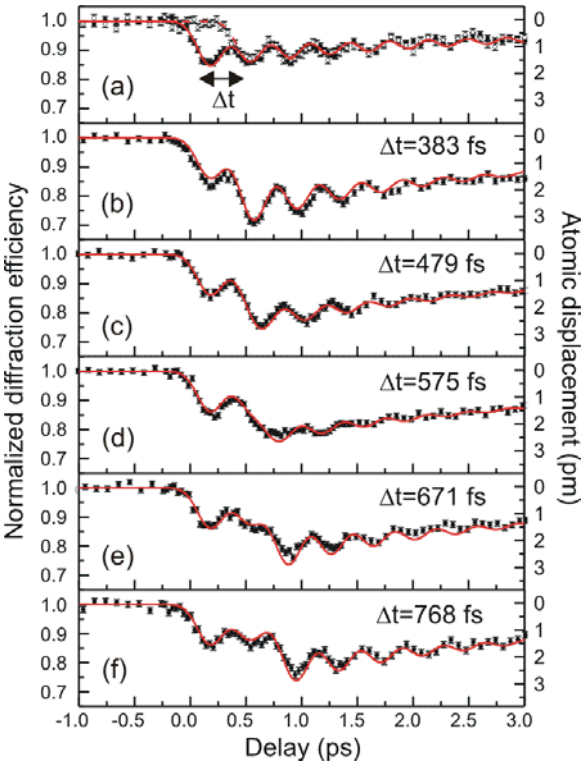

Figure 2: *Measured (symbols) and simulated (solid curves) bismuth (111) coherent optical phonon oscillation controlled by a 2-pulse excitation scheme. The absorbed fluence per pulse was 1.12 mJ/cm^2. Right axis: Atomic displacement as inferred from the simulation.*

To demonstrate the reliability of the source to perform systematic measurements over a longer time period, we used a sequence of two pulses to control the population of the A_{1g} phonon mode. In this double-pump experiment, the amplitude of the coherent atomic motion is manipulated through the delay of the second pulse [3]. The spectra shown in Fig. 2(b-f) are obtained by delaying the second pump pulse over one oscillation period of the coherent optical phonon. Its amplitude is either amplified [Figs. 2(b) and 2(f)] or canceled [Fig. 2(d)], depending on the relative phase of the second pulse with respect to the coherent phonon generated by the first pulse. The simulated curves are obtained using the atomic potential of Ref. [3]. Figure 2 demonstrates to the best of our knowledge the first optical control experiment were the coherent structural response has been probed directly with hard x-rays with femtosecond resolution.

References

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Publications

- **Spatiotemporal stability of femtosecond hard x - ray undulator radiation studied by control of coherent optical phonons**
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  Phys. Rev. Lett. 99, 174801 (2007)
  doi: 10.1103/PhysRevLett.99.174801

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