Picosecond Time-resolved X-ray Diffraction at the APS Sector 7 (MHATT-CAT)

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Introduction

Sub-picosecond x-rays can be used to resolve motion on the time scale of molecular dynamics. This is considerably shorter than the typical 50–100 ps pulse duration from a synchrotron. There have been a number of recent attempts to produce ultra-fast x-rays using intense laser radiation including K_α radiation from laser induced plasmas [1], Thompson scattering of a laser pulse from a relativistic electron beam [2], ponderomotive scattering of the electron beam from a laser pulse in a synchrotron [3], and the interruption of diffraction from a Bragg mirror with rapid heating from a short laser pulse [4]. Fourth generation (free-electron laser based) sources will produce ultra short bursts of x-rays from the electron bunches from a linear accelerator [5].

Another method to produce ultrafast x-ray pulses (that can be used with existing storage ring based sources) is to create a laser induced superlattice on a Bragg reflector [6]. This superimposed periodicity changes the Bragg condition on an ultrafast time scale and can be coherently controlled to produce a switch that can be turned on and off at integer half multiples of the excited optical phonon frequency (approximately 100 fs in GaAs). An alternative but closely related method is to excite near zero wave vector coherent optical phonons which can create allowed reflections from forbidden ones [7]. These are the approaches that are being used to produce the ultrafast x-ray pulses for use on the MHATT-CAT beamline of the Advanced Photon Source.

Experimental Approach

A commercially available Ti:sapphire 840 nm laser has been installed on the insertion device beam line (7ID-D). The laser is capable of impulsively driving large amplitude acoustic and/or optical phonon fields in a variety of materials. The front end of the laser consists of a Ti:sapphire oscillator which is phase-locked to the 352 MHz of the accelerator cavities in the storage ring. Due to phase noise in the delivered rf, the jitter between the oscillator and the rf is on the order of a 20 ps (rms). The jitter between the laser and the electron beam (and thus the x-rays) is believed to be less than 70 ps; however, a future upgrade to the timing reference is expected to reduce this to < 5 ps. The amplified laser pulses are delivered at a 1 kHz repetition rate with up to 0.75 mJ/pulse in a pulse as short as 50 fs and are synchronized with a single bunch in the storage ring.

The x-rays are collimated 26.5 meters from the undulator with 400 x 400 μm slits before being monochromatized by a cryogenically cooled Si (111) double crystal monochromator. The energy was set at 10 keV with a width of approximately 1.4 eV. At approximately 58 meters from the source, the beam is again collimated with 100 x 800 μm slits less than a meter from the sample. The flux is monitored with an ionization chamber after the slit and is ~ 5 10^{10} photons/s/ 100 mA which corresponds to 7 10^{3} photons/ bunch/ 100 mA. The Bragg reflectivity of the InSb sample is > 20%. A silicon avalanche photodiode (APD) with rise time of 3 ns was used for the time-resolved detector.

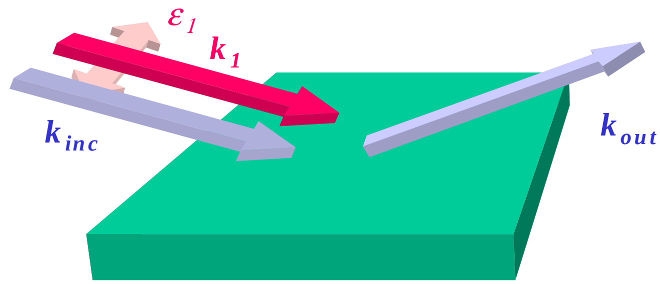

Figure 1. Geometry for ultrafast x-ray scattering. k_1 is the incident laser and k_{inc} (k_{out}) are the incident (reflected) x-rays.

Results

The first step in commissioning the ultra-fast dynamics capability has been to study rapid disordering in InSb. The laser is focused to a roughly 1 x 5 mm spot upon the InSb at near grazing incidence to the (111) surface in a direction roughly parallel to the incident x-rays (Figure 1.). Figure 2. shows the time-resolved rocking curve of a single 10 kV x-ray pulse diffracting from the (111) surface of the InSb crystal when stimulated by the laser just below the InSb damage threshold. The time was scanned by delaying the laser pulse with respect to the single-bunch x-ray pulse with 19 ps resolution. Because the APD is relatively slow, we measure the convolution of the x-ray pulse duration (as well as the timing jitter) at each temporal location. Nonetheless, many interesting time-dependent features persist. The data seem consistent with an impulsively generated strain

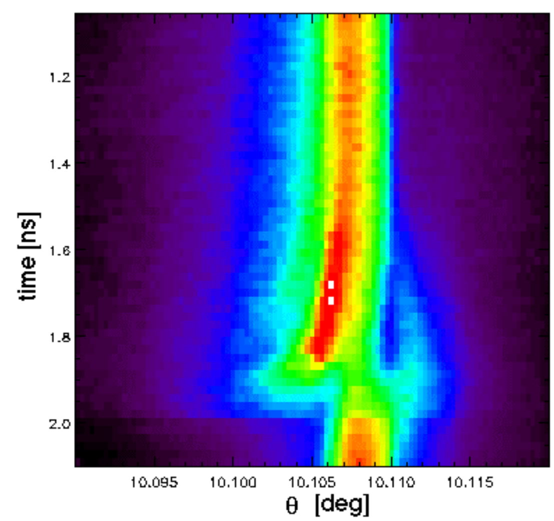

Figure 2. Time-resolved rocking curve of the (111) reflection of InSb impulsively heated by an 840 nm laser pulse. The laser arrives later than the x-rays at longer times (towards the bottom of the figure).

that propagates into the crystal at the speed of sound [1,4]. At angles larger than the Bragg angle for the unperturbed sample, one can see the effect of the compression side of the strain wave, which persists until the wave has penetrated past the extinction depth of the x-rays. Near the Bragg peak where the phonons are slow, one can resolve coherent oscillations. This is even more evident on the rarefaction side of the strain wave (smaller angles). This area of lattice expansion is larger and closer to the surface, and thus diffracts more x-rays. In addition one sees that the whole of the rocking curve shifts towards smaller angles due to the average increase in the lattice constant. This shift then slowly decays on the many nanoseconds time scale.

Discussion

A major milestone in the commissioning of the of the MHATT-CAT insertion device beamline has been the observation of time-resolved x-ray scattering from coherent acoustic phonons in InSb. Experiments are continuing towards the development of an ultrafast x-ray switch which utilizes coherent optical phonons (these are much faster than their acoustic counterparts). Coherent control over x-rays diffraction on the sub-picosecond time scale will provide a valuable tool for the studies of ultrafast lattice dynamics.

Acknowledgments

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