Ultrashort Flashes of X-rays and Electrons at SLAC

Since the development of the laser researchers have been using it as a stroboscopic tool to observe how the world works. Research and development efforts leading towards the Linac Coherent Light Source (LCLS) free-electron laser have facilitated the construction of a new accelerator-based femtosecond x-ray source, the Sub-Picosecond Pulse Source (SPPS) which extends our ability to capture transient phenomena at atomic-scale resolution. In order to produce femtosecond x-ray bursts, electron bunches at SLAC are chirped and then sent through a series of energy-dispersive magnetic chicanes, yielding 80-fs electron pulses. These are then transported through an undulator to create sub-100-femtosecond x-ray pulses at 8-10 keV and 10^7 photons/second.

Two recent experiments[1,2] have now used this source to elucidate the ultrafast transition from solid to liquid, and demonstrated a method to both measure the duration of a femtosecond electron bunch and to overcome the jitter between a linac-based x-ray source and a femtosecond laser.

The solid-liquid transition

Intense femtosecond excitation of semiconductor materials results in the excitation of a dense electron-hole plasma, with accompanying dramatic changes in the interatomic potential, leading to disordering of the material on timescales shorter than electron-phonon coupling times. Pump-probe measurements using an InSb sample were conducted in a cross-beam geometry with the optical pump pulse incident at an angle with respect to the x-ray probe pulse. In this way, a temporal sweep is created along the crystal surface, transforming temporal information into spatial information and enabling one to record the complete time history around t=0 in a single shot.

Figure 1 shows the measured time-dependent intensity I(Q,t) for both the (111) and (220) reflections, averaged over 10 single shot images. We observe that the diffracted intensity is non-

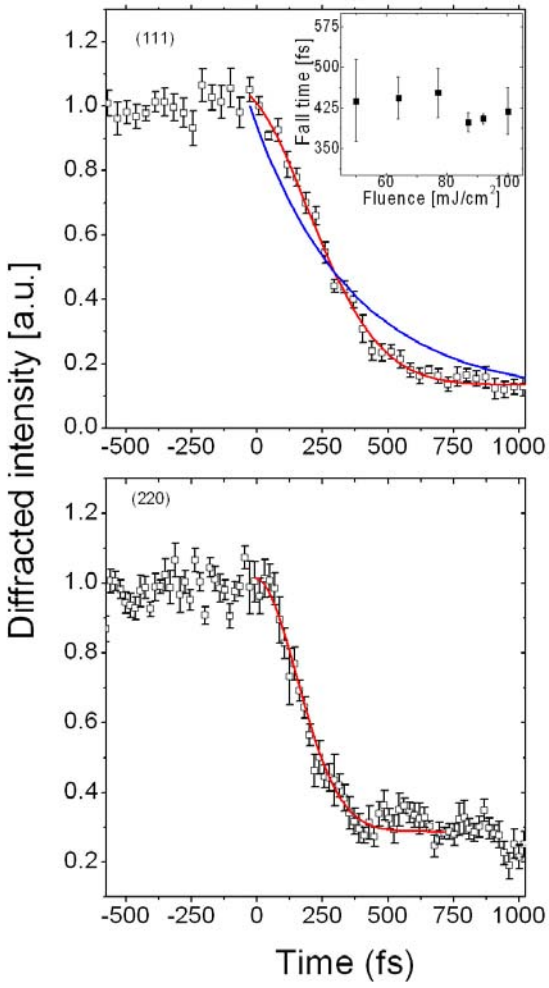

Fig. 1. (top) Time-resolved diffracted intensity for both (111) (top) and (220) (bottom) reflections. Red curves are Gaussian fits to the data, corresponding to 10-90 % fall times of 430 fs and 280 fs respectively. (inset) Fluence dependence of (111) time constant.

exponential and well-fit by a Gaussian-like curve. Moreover, the (220) reflection decays with a time-constant qualitatively faster than the (111). The 10-90 time constants for the (111) and (220) reflections are 430 fs and 280 fs respectively, with ratio $\tau_{(111)} / \tau_{(220)} = 1.5 \pm 0.2$. This is equal (within experimental error) to the ratio of the magnitude of the reciprocal lattice vectors for the two reflections. This inverse-Q-dependent scaling and Gaussian-like time-dependence strongly implies statistical atomic motion and suggests that the data be described using a time-dependent Debye-Waller model, relating the time-dependent decrease in scattered intensity to a time-dependent rms displacement, as $I(Q,t) = e^{-2W} = e^{-\frac{Q^2 <u^2(t)>}{3}}$. Here <u^2(t)> is the time-dependent mean square displacement of the photo-excited atoms, averaged spatially over the sample. Following an impulsive softening of the interatomic potential, for short times afterwards (to first order in t) the atoms continue to move with velocities set by initial conditions, i.e. inertially, following Newton's First Law. The time-resolved diffracted intensity I(Q,t) is then expected to be Gaussian in both Q and t with a time-constant that varies inversely with Q, exactly as observed. By inverting the time-dependent data one may extract the time dependence of the rms displacement. We measure a linear time-dependence which corresponds to a velocity 2.3 *Å/ ps* in good agreement with the room temperature rms velocity in InSb $(3k_bT/M)^{1/2}$ = 2.5 *Å/ ps*. The implication is then of a transition state in which the potential energy is softened enough that the atoms initially move freely with large amplitude along an effectively barrierless potential energy surface with initial conditions set by room-temperature thermodynamic velocities. During the first few hundred fs, although the displacements correspond to a large fraction of the interatomic distance and the strong covalent bonds characteristic of the crystalline state are broken or strongly modified, the average displacement <u> from the equilibrium lattice sites is still zero, a state intermediate between that of a solid and a liquid.

Clocking femtosecond x-rays

A non-invasive technique is used to record the arrival time of femtosecond electron bunches at SPPS with respect to a pump laser pulse. Although the time of arrival fluctuates from pulse to pulse, this information can be used to place repetitive measurements in sequence with femtosecond resolution. The optical properties of an electro-optic crystal placed adjacent to the electron beam are strongly modified as a result of the electric-field of the electron bunch as it passes. A femtosecond laser pulse propagating through the crystal at the same time as the transient birefringence is induced has its polarization rotated and can thus be used as a probe of the relative arrival time of the electron bunch. By making use of a cross-beam geometry like the melting experiment described above, a range of times is measured, and the timing information is imprinted on the spatial profile of the transmitted laser beam.

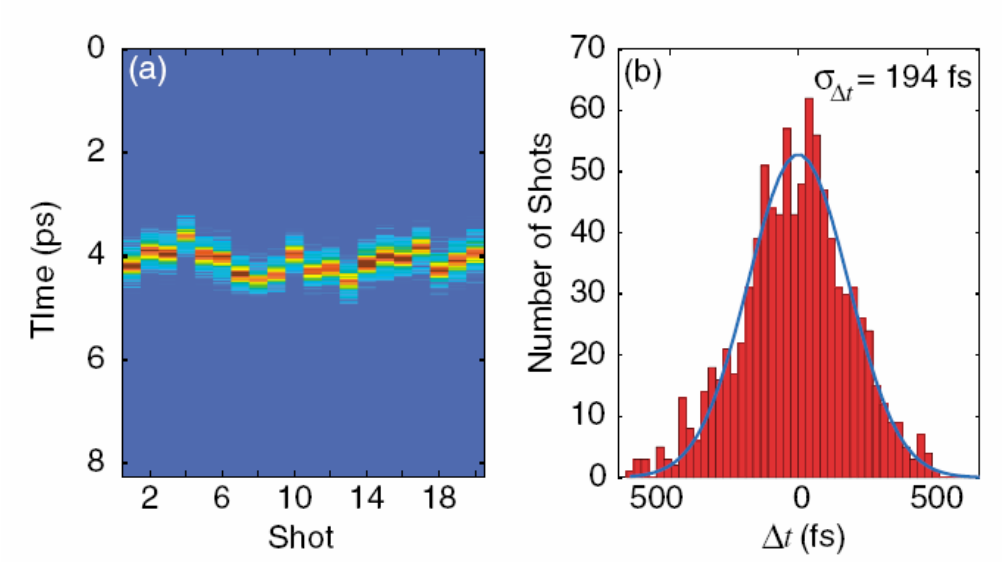

Fig. 2. (a) 20 consecutive single shot electron bunch measurements. The bright band in each column is the electro-optic signal, its location indicates the time of arrival of the electron bunch with respect to the laser probe pulse, and its width corresponds to the electron bunch duration. b) Normalized arrival time histogram of 1000 consecutive single shots.

This experiment makes possible a wide range of repetitive measurements with time-resolution limited by the pump and probe durations instead of the intrinsic jitter between pump and probe. Relative timing information from spatially-resolved electro-optic measurements could be extended to a resolution of order 5 fs, matching the projected performance of future XFELs into the foreseeable future.

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