

Condensed Matter Highlight

A film of picosecond vibrations of a film

David A. Reis*

FOCUS Center and Department of Physics, University of Michigan, 450 Church Street, Ann Arbor, MI 48109 1040, USA

Received 19 September 2005; accepted 21 September 2005 by R. Merlin

Available online 3 October 2005

Time-domain experiments on solid-state dynamics at the fastest time-scales are typically performed using all optical techniques. These experiments have been made possible by, and have driven significant advances in, pulsed laser technology. Since optical techniques only indirectly probe structure, there is a significant scientific motivation for using higher spatial resolution probes such as X-ray and electron diffraction or microscopy. Unlike lasers, ultrafast X-ray and electron sources are much less developed. Nonetheless, there has been significant advances in ultrafast science using short wavelength probes, showing a similar synergy between source and science as was seen early on in the laser-based community. For their part, electrons are much more strongly interacting with matter than X-rays (the Coulomb is much larger than the Thompson cross-section) and are particularly well suited to studies in the gas phase [1]. Despite the large cross-section for elastic scattering from nuclei, many electrons are required to make a diffraction image, and space charge forces make it difficult to deliver these pulses in a short burst. In the solid phase, most of the (short pulse short wavelength) experiments to date has centered around X-ray diffraction. Here synchrotron based sources provide very bright and very flexible beams with roughly 100 ps duration [2,3], while table top laser-plasma based sources provide subpicosecond duration pulses albeit with very low brightness (but enough photons for single crystal diffraction [4,5]). Nonetheless, time-resolved electron diffraction is a complementary technique that is showing tremendous promise for ultrafast solid-state dynamics.

One such experiment is reported in this issue in the article by Park et al. [6]. They report on their success in imaging the coherent atomic motion in a thin metallic film that has been set vibrating by a femtosecond laser pulse. In

their experiments, the laser pulse excites a single longitudinal breathing mode, a standing wave, with a wavelength of twice the thickness of the aluminum film or only 40 nm. At this wavelength the film thickness oscillates with a period of just over 6 ps. Images of the oscillating film are obtained with very high spatial and temporal resolution using a home-built time-resolved electron diffraction apparatus. In the past, other groups have used time-resolved electron diffraction to study large scale changes in structure during melting, first with tens of picoseconds resolution [7] and, more recently, subpicosecond resolution [8]. In contrast, the experiments reported in this issue concentrate on relatively subtle changes of the structure. While the relative changes to the diffraction patterns are small, a fraction of a percent, high contrast images were recorded with sufficient resolution to freeze the atomic motion.

To view the dynamics, the authors generate short bursts of electrons with kinetic energy of 60 keV and shoot them through the polycrystalline film. A fraction of these electrons diffract from a collection of crystal grains, and their pattern is collected to give a subpicosecond snapshot of the structure of the film (the film is thin enough that multiple scattering can be neglected). The experiments are table-top scale with the electrons derived from a photocathode using a fraction of the same laser pulse that excited motion in the film. In this manner, a movie of the dynamics (a film of the film) was recorded in a stroboscopic, pump-probe fashion, by adjusting the time delay between the laser (pump) and the electron beam (probe) with a variable optical delay line. The authors looked in this movie for time-dependent changes in radius of the diffraction rings, corresponding to the changes in the spacing and orientation of the crystalline planes. They found that the radius of each diffraction ring oscillated with the period of the coherent acoustic phonons corresponding to the standing wave, and that each ring was in phase with the other and oscillated about a new equilibrium position in

* Tel.: +1 734 763 9649; fax: +1 734 764 5153.

E-mail address: dreis@umich.edu.

a damped cosine-like fashion. This is indicative of a displacive type excitation where the atoms are sent into motion not by a direct impulse, but instead by finding themselves suddenly under the influence of a new binding force, due to the nonthermal photoexcited electrons, and thus a new equilibrium position (consider as an oversimplification, the difference between pushing a child on a swing and suddenly moving the swingset). In addition to the coherent motion, Park et al. also observe the time required for lattice thermalization (electron–phonon coupling). This is measured through changes in the incoherent thermal vibrations of the ions, which decrease the intensity of the diffraction rings with increasing temperature. The time-constant for thermalization was subpicosecond such that the equilibrium temperature was reached within roughly a quarter of the coherent phonon period. Importantly, this is several times larger than would be predicted using a two temperature model for the electrons and ions, and suggest that more detailed studies are sure to follow.

These experiments demonstrate the potential for ultrafast electron diffraction as a sensitive probe of structural dynamics, in a laboratory setting. We note that in the hard X-ray realm much of the source development is geared towards linear accelerators such as the 3 km Stanford linear accelerator, where sub 100 fs pulses with peak brightness exceeding the third generation storage ring are currently being used [9] and another 10 orders of magnitude (and the potential for attosecond pulses) is expected with the construction of the linac coherent light source, X-ray free electron laser [10]. Will time-resolved electron diffraction follow a similar path? There are a number of trade offs [11]. For example, it is easier to transport short electron pulses if they are relativistic, but, for a given set of planes, the decrease in the diffraction angle requires a long drift. However, accelerator-based sources lack the intrinsic synchronization afforded to table top sources [12]. Although the momentum transfer is the same, the electrons become much more penetrating. Of course, if one were to go to thicker samples, multiple scattering would become an issue. As in the case of X-ray diffraction, this can also be advantageous [13]. In the end, whether its electrons or

X-rays, table-top or national user facility, all these techniques are complementary and pushing the frontiers of time-domain science.

References

- [1] R. Srinivasan, V.A. Lobastov, C.-Y. Ruan, A.H. Zewail, Ultrafast electron diffraction (UED) a new development for the 4D determination of transient molecular structures, *Helv. Chim. Acta* 86 (2003) 1763–1838.
- [2] A.M. Lindenberg, et al., Time-resolved X-ray diffraction from coherent phonons during a laser-induced phase transition, *Phys. Rev. Lett.* 84 (2000) 111–114.
- [3] D.A. Reis, et al., Probing impulsive strain propagation with X-ray pulses, *Phys. Rev. Lett.* 86 (14) (2001) 3072–3075.
- [4] C. Rose-Petruck, et al., Picosecond–milliangstrom lattice dynamics measured by ultrafast X-ray diffraction, *Nature* 398 (1999) 310–313.
- [5] K. Sokolowski-Tinten, et al., Femtosecond X-ray measurement of coherent lattice vibrations near the Lindemann stability limit, *Nature* 422 (2003) 287–289.
- [6] H. Park, X. Wang, S. Nie, R. Clinite, J. Cao, Direct and real-time probing of both coherent and thermal lattice motions, *Solid State Commun.* this issue (doi: 10.1016/j.ssc.2005.07.034).
- [7] S. Williamson, G. Mourou, J.C.M. Li, Time-resolved laser-induced phase transformation in aluminum, *Phys. Rev. Lett.* 52 (6) (1984) 2364–2367.
- [8] B.J. Siwick, J.R. Dwyer, R.E. Jordan, R.J.D. Miller, An atomic-level view of melting using femtosecond electron diffraction, *Science* 302 (2003) 1382–1385.
- [9] A.M. Lindenberg, et al., Atomic-scale visualization of inertial dynamics, *Science* 308 (2005) 392–395.
- [10] LCLS conceptual design report, SLAC-R-593 (2002).
- [11] W.E. King, et al., Ultrafast electron microscopy in materials science, biology, and chemistry, *J. Appl. Phys.* 97 (2005) 111101.
- [12] A.L. Cavalieri, et al., Clocking femtosecond X-rays, *Phys. Rev. Lett.* 94 (2005) 114801.
- [13] M.F. DeCamp, et al., Transient strain driven by a dense electron–hole plasma, *Phys. Rev. Lett.* 91 (16) (2003) 165502.