Lattice Motions from THz Phonon-polaritons measured with Femtosecond X-ray Diffraction.

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Abstract. We use femtosecond x-ray diffraction to measure the coherent lattice displacements associated with the excitation and propagation of THz phonon polaritons in $LiTaO_3$.

Phonon-polaritons are light waves propagating within a polar solid at frequencies near one or more vibrational resonances. They give a microscopic description of the index of refraction and their study is particularly important in the THz region of the spectrum, where strong coupling between light and lattice is found and where electromagnetic energy transport has important ramifications in both fundamental science and technology [1]. Femtosecond optics has opened new opportunities in this area, making it possible to excite coherent phonon polaritons impulsively [2], to detect their amplitude and phase in the time domain, and to manipulate their spatial and temporal properties [3]. Yet, because of the lack of a suitably fast structural probe, all experiments to date have concentrated on the measurement of the time-dependent electrical polarization, while the corresponding lattice displacements have remained undetected. Here, we apply femtosecond x-ray diffraction to directly measure atomic motions associated with 1.5-THz, coherent phonon polaritons of Ag symmetry in ferroelectric $LiTaO_3$.
Optical-pump and x-ray-probe pulses were made to impinge collinearly at 68 degrees from the c axis of the crystal. This angle, which was dictated by the 006 Bragg condition for 7 keV x-rays, was chosen also for the optical pump, to maximize temporal resolution. The 10-mJ/cm^2, 800-nm pulse, focused onto a 1-mm spot size, was refracted into $LiTaO_3$ at 25 degrees from the surface normal, while phonon polaritons were excited by Impulsive Stimulated Raman Scattering at the surface of the crystal. Due to the high index of refraction at THz frequencies (n~6), surface-generated phonon polaritons propagated into the crystal at 8 degrees from the surface normal and at lower phase velocity then the optical pump. Figure 1 shows a simulation of the generation and propagation of these polariton waves.

Femtosecond x-ray diffraction measurements were performed at beamline 5.3.1. of the LBNL Advanced Light Source, where laser modulation of the electron beam energy [5] was exploited to generate femtosecond pulses of bending magnet radiation. Such pulses were synchronized to an external laser and were continuously tunable between the visible and the hard x-rays, allowing for femtosecond x-ray experiments over a broad spectrum [6].

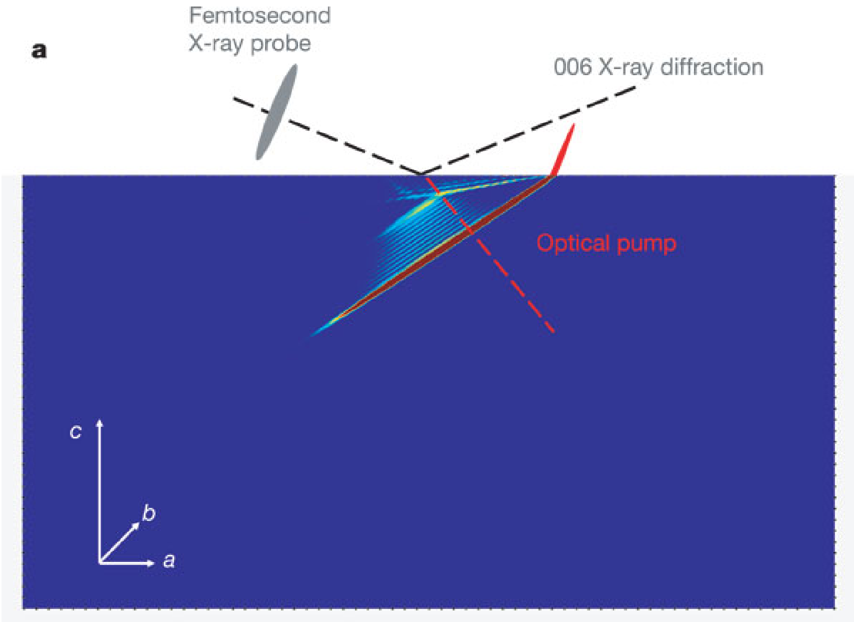

Figure 1: Simulation of polariton excitation and propagation in $LiTaO_3$ and geometry of femtosecond x-ray diffraction experiment.

The 006 reflection was chosen because it has the most sensitive structure factor to the Ag displacements along the z axis. Because h=k=0, these time-resolved 006 x-ray diffraction cannot detect distortions in the x-y plane (E mode). Due to strong scattering in the 006 direction, the x-ray experiments were only sensitive to the first 1 micron beneath the surface.

Our measurements are shown in figure 2, where oscillations at nearly 1.5 THz are clearly identified. The figure also displays a calculation of the time-dependent 006 structure factor, as derived from the simulations of figure 1, a calculation of the atomic motions associated with the Ag normal mode, and the resulting time-dependent diffraction in kinematic approximation. Only the amplitude of the simulated curve was scaled to fit the data. Good agreement is found.

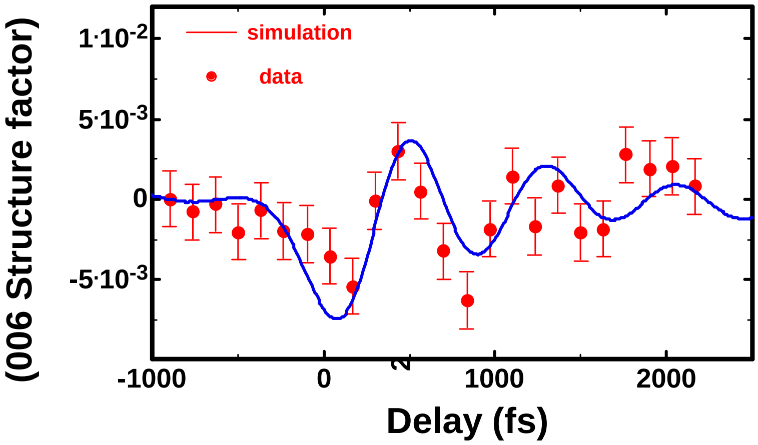

Figure 2: Time-resolved x-ray diffraction measurements and simulations of the time-dependent 006 reflection in LiTaO3.

In conclusion, we have used femtosecond x-ray diffraction to measure coherent atomic displacements associated with the excitation and propagation of a near-1.5-THz phonon polariton in $LiTaO_3$[6].

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