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Dynamical Studies of Phonon Polaritons by Femtosecond X-ray Diffraction MATTEO RINI, ROBERT W SCHOENLEIN, Lawrence Berkeley National Lab, ERIC STATZ, DAVID W WARD, KEITH A NELSON, Massachusetts Institute of Technology, SIMON WALL, CHRIS SIMPSON, ANDREA CAVALLERI, Department of Physics, University of Oxford — In solids, light propagation near resonances can be described in terms of polaritons, mixed excitations with both phonon- and photon-like character arising from the coupling of the oscillating electromagnetic field to polar lattice vibrations. Here we use time-resolved x-ray diffraction with 200-fs synchrotron pulses to directly measure the coherent lattice distortions associated with the propagation of THz phonon polaritons in ferroelectric $LiTaO_3$. THz radiation was generated by fs IR pulses via impulsive stimulated Raman scattering. The lattice motion component of polariton propagation was measured as time-dependent modulation of the 006 structure factor. The comparison with a time-domain simulation of THz propagation allowed extracting the absolute atomic displacements along the coordinate of the A_1 normal mode with 1-mÅ resolution. Our experiments reveal a polariton wave with a 1.5-THz frequency and a 5-mÅ peak displacement of the Ta atom with respect to the plane of the oxygen atoms. Complementing optical techniques which measure the time-dependent electrical polarization, femtosecond X-ray experiments directly monitor amplitude and phase of all structural degrees of freedom.

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