Coherent control of phonons probed by time-resolved x-ray diffraction

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Time-resolved x-ray diffraction with picosecond temporal resolution is used to probe the product state of a coherent control experiment in which a single acoustic mode in a bulk semiconductor is driven to large amplitude or canceled out. It is demonstrated that by shaping ultrafast acoustic pulses one can coherently control the x-ray diffraction efficiency of a crystal on the time scale of a vibrational period, with application to coherent switching of x-ray beams. © 2002 Optical Society of America

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By making use of classical or quantum-mechanical interferences, one can use light to control the temporal evolution of atomic, molecular, and solid-state systems and to drive them into novel, nonequilibrium states of matter. For example, appropriately timed sequences of femtosecond light pulses maximize the yield of a chemical reaction[1] or selectively excite a vibrational mode in a solid.[2] The resulting product states have until now been probed by indirect means, using light with a wavelength much larger than an interatomic spacing. Here we use time-resolved x-ray diffraction[3–5] to measure, with atomic-scale spatial resolution, the coherently controlled structural dynamics following two-pulse femtosecond excitation of a bulk solid. It is shown how particular bulk acoustic phonon modes can be selectively enhanced or completely canceled out.

Although coherent control of acoustic modes was demonstrated previously using optical pump–probe techniques,[6–8] this has not been achieved in a bulk material. This is due to the difficulty of using visible-light techniques to probe large wave-vector modes. Recent experiments have thus focused on multiple-quantum-well structures or semiconductor superlattices, materials that exhibit nonzero-frequency acoustic modes at the Brillouin zone center. In contrast, x rays, with wavelength comparable to atomic spacings allow one to measure the entire Brillouin zone, in principle up to wave vectors of the order of an inverse lattice constant.

In the experiment described here, the vibrational modes of the solid are probed by time-resolving the inelastic scattering of x rays by phonons. In the frequency domain, inelastic x-ray scattering[9] has been used to probe vibrational excitations by resolving the change in energy of a photon scattered by a phonon at a given momentum transfer q. Alternatively,[3] time-resolved x-ray diffraction can be used to observe directly the vibrational motion associated with a particular phonon mode. A phonon of wave vector q modulates the diffraction efficiency of a crystal at a corresponding frequency ω. By resolving this effect on time scales comparable to vibrational periods, one can directly record the phonon frequency corresponding to a given q. With sufficient temporal resolution, the scattering off milli electron volt–energy-scale acoustic phonons can be resolved using x rays with electron volt–scale bandwidth.

The phonons probed in this experiment are excited impulsively through femtosecond optical excitation.[10] Initial energy deposition on a time scale faster than the lattice can respond results in the buildup of multikilobar pressure at the surface. This stress is then released by an acoustic pulse, consisting of a region of compression followed by a region of expansion, which propagates into the solid at the velocity of sound. The generated acoustic strain profile after 60 ps is shown in the inset of Fig. 1. The width of this pulse is of the order of the laser penetration depth (100 nm) and corresponds to a broad spectrum of excited modes of width $\sim 10^5$ cm^{-1}.[10] Superimposed on this coherent pulse is an incoherent part corresponding to an expansion near the surface of the crystal. Whereas the coherent component propagates

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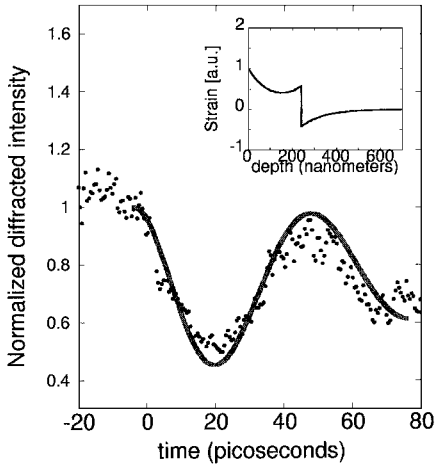

Fig. 1. Single-pulse excitation, probing the $q = 4 \times 10^5$ cm^{-1} mode. Solid curve, numerical simulation. Inset: induced strain profile at time $t = 60$ ps.

ballistically and leaves the probed region within ~100 ps, the incoherent part propagates diffusively and lasts for hundreds of nanoseconds.

Two-pulse excitation impulsively excites a particular acoustic mode twice, the second time with some chosen phase relative to the first. The first pulse starts a coherent motion with all atoms moving in synchrony. The second excites a phase-delayed replica of the first, and the two interfere constructively or destructively. The final product state is a system with one selected mode excited to large amplitude or canceled out. Equivalently, one may view two-pulse excitation as pulse shaping of an acoustic pulse, analogous to the shaping of femtosecond optical pulses used in a variety of coherent control schemes.[11,12] From the standpoint of controlling the x-ray diffraction efficiency of a crystal, shaped acoustic pulses are being used to define the local amount of order within a crystalline solid.

The experimental setup has been described elsewhere.[3] X rays at the Advanced Light Source synchrotron are monochromatized to an energy of 5 keV with a spectral bandwidth of 3 eV. A Ti:Al_2O_3-based 150-fs, 1-kHz, 800-nm laser is synchronized to individual electron bunches within the storage ring with 5-ps jitter. A Michelson interferometer splits the laser into two pulses. The incident fluence for each pulse is 2 mJ/cm^2. The time-resolved diffracted intensity is then measured with a streak camera with 2-ps resolution in averaging mode.[13,14] In this way we obtain the complete time history in a 100-ps window following excitation. The sample is bulk crystalline InSb, cut asymmetrically so that the [111] planes are oriented at an angle of 16° from the surface and used at grazing incidence. In this geometry the dephasing time is set by the time it takes the generated acoustic pulse to leave the x-ray probe region, rather than by the bandwidth of the monochromator or the intrinsic divergence of the synchrotron source. The generated acoustic pulses thus exhibit coherences that last of the order of 100 ps.

Figure 1 shows the diffracted intensity (normalized to 1 for $t \leq 0$) for single-pulse excitation at an angle 300 arcsec off the (111) Bragg reflection, corresponding to the $q = 4 \times 10^5$ cm^{-1} mode. Following an initial drop in the diffracted intensity, large amplitude oscillations with period ~40 ps are observed, corresponding to the selected mode according to the linear dispersion relation $w = qv_{\text{sound}}$ ($v_{\text{sound}} = 4000$ m/s). This result can be qualitatively understood in the following way: Nonuniform deposition of heat on a time scale faster than the acoustic vibrational period generates new energetically favorable displacements. The lattice then moves toward this new equilibrium state but overshoots it and undergoes oscillations about the new equilibrium position. This is the essence of a displacive excitation, as more typically applied to the case of optical phonon excitation.[16] Thus, oscillations in the measured diffracted intensity are induced about a constant value corresponding to the new equilibrium lattice displacement, as qualitatively observed in Fig. 1, for single-pulse excitation.

Two-pulse excitation creates acoustic pulses of the form shown in the insets of Fig. 2. Note that at the excitation powers used in this study the phonon amplitude depends linearly on the excitation fluence. Phonon–phonon interactions are furthermore negligible. Thus the acoustic pulses obey linear superposition. The inset of Fig. 2a shows the acoustic pulse generated by two-pulse excitation with a delay of 35 ps, equal to one vibrational period. This profile drives the $q = 4 \times 10^5$ cm^{-1} mode to large amplitude and suppresses all nearby modes, as shown in Fig. 2a. Comparison with the single-pulse data in Fig. 1 shows that the vibrational amplitude builds up over time, a result of constructive interference. In contrast (Fig. 2b), for a relative delay of 18 ps, or 1/2 a vibrational period, the probed mode is completely silenced. The insets to Figs. 2a and 2b thus represent the shape of the optimal acoustic pulses for constructive or deconstructive interference of the $q = 4 \times 10^5$ cm^{-1} mode (for nonzero pulse delays). Note that the oscillation frequency differs slightly between the one- and two-pulse excitation cases. This is a result of a larger shift of the Bragg peak to smaller angles for the two-pulse case, leading to a slightly larger probed phonon wave vector for the two-pulse case.

Also shown in Figs. 1 and 2 are simulations of the time-resolved diffracted intensity. These correspond to solutions of the x-ray dynamic diffraction equations[17] with the strain fields shown in the insets. We extract a maximum amplitude of ~0.03 nm for the coherently enhanced mode. In contrast, for the case of deconstructive interference, the amplitude of the controlled mode is zero within our signal-to-noise ratio.

We note that the ability to completely cancel sidebands on x-ray reflections has application in the development of ultrafast x-ray switches[18] capable of slicing out a portion of the long x-ray pulses typically generated at synchrotrons. Switching of synchrotron x-ray pulses on a 100-ps time-scale was recently reported.[19] In this work, switch-off times of the order of 15 ps, significantly faster than the duration of synchrotron x-ray pulses, are generated with a loss factor of 10 in intensity. Significantly

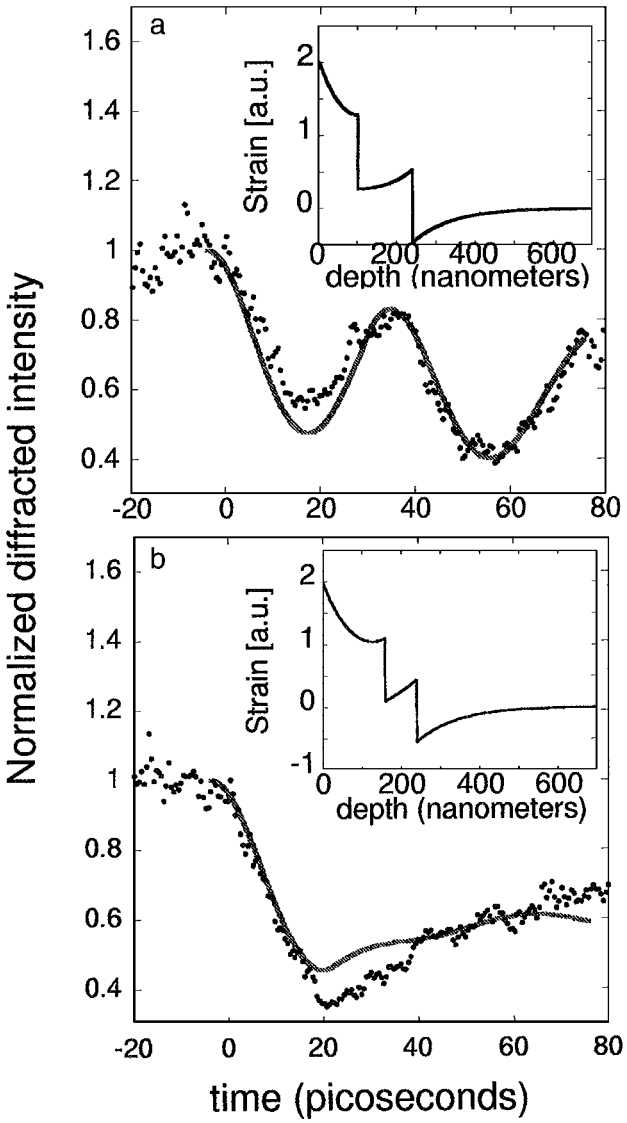

Fig. 2. a, Enhancement of the $q = 4 \times 10^5$ cm^{-1} acoustic mode by two-pulse excitation with a relative delay of 35 ps, equal to one vibrational period. b, Cancelation of the $q = 4 \times 10^5$ cm^{-1} mode by two-pulse excitation with a relative delay of 18 ps, or 1/2 a vibrational period. Solid curves, numerical simulations. Insets: optimal strain profiles for constructive or destructive interference of the $q = 4 \times 10^5$ cm^{-1} mode.

faster switching times are possible by tuning the angle of the crystal to select higher wave-vector modes (with larger associated losses).

In summary, time-resolved x-ray diffraction techniques have been used to observe directly the coherent excitation and control of selected acoustic vibrational modes in a bulk semiconductor. In the future the use of time-resolved x-ray scattering techniques to probe the coherently controlled dynamics of matter on an atomic length scale should lead to the optimization of more-complicated processes (for example, the creation of new structural phases of matter), in which the optimal control sequence is not yet known but may be extracted through the direct observation, in real time, of the resulting atomic motion.[20]

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