Supersonic strain front driven by a dense electron-hole plasma

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We study coherent strain in (001) Ge generated by an ultrafast laser-initiated high density electron-hole plasma. The resultant coherent pulse is probed by time-resolved x-ray diffraction through changes in the anomalous transmission. The acoustic pulse front is driven by ambipolar diffusion of the electron-hole plasma and propagates into the crystal at supersonic speeds. Simulations of the strain including electron-phonon coupling, modified by carrier diffusion and Auger recombination, are in good agreement with the observed dynamics.

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Subpicosecond laser-induced electron-hole plasmas in semiconductors can produce large amplitude lattice strain and rapid loss of translational order. These effects have been studied extensively in ultrafast linear and nonlinear reflectivity experiments and, more recently, in time-resolved x-ray Bragg scattering experiments. X-ray diffraction has the advantage that it can provide quantitative structural information. Many of the x-ray experiments have been analyzed using the thermoelastic model put forth by Thomsen et al. in which the strain is caused by differential thermal expansion. Deviations from this model are discussed in the work of Thomsen et al. and have been seen in x-ray diffraction and, more recently, in time-resolved x-ray Bragg scattering experiments. X-ray diffraction has the advantage that it can provide quantitative structural information. Many of the x-ray experiments have been analyzed using the thermoelastic model put forward by Thomsen et al. in which the strain is caused by differential thermal expansion. Deviations from this model are discussed in the work of Thomsen et al. and have been seen in x-ray diffraction and, more recently, in time-resolved x-ray Bragg scattering experiments. X-ray diffraction has the advantage that it can provide quantitative structural information. Many of the x-ray experiments have been analyzed using the thermoelastic model put forward by Thomsen et al. in which the strain is caused by differential thermal expansion. Deviations from this model are discussed in the work of Thomsen et al. and have been seen in x-ray diffraction and, more recently, in time-resolved x-ray Bragg scattering experiments. X-ray diffraction has the advantage that it can provide quantitative structural information.

In this letter we report on measurements of coherent strain generation in Ge following ultrafast laser-excitation using a bulk sensitive structural probe. We use time-resolved ultrafast x-ray transmission to measure strain propagation deep within the crystal, providing information about the generation process. Initially, the strain front advances at speeds greater than the sound speed. In our experiments, the laser intensity is sufficient to impulsively generate a dense electron-hole plasma at the crystal surface, the dynamics of which are governed by ambipolar diffusion and Auger recombination. The plasma couples to the lattice through the deformation potential. In order to probe the resulting coherent acoustic pulse as it travels deep within the bulk, we utilize the Laue geometry whereby the x-rays traverse the full thickness of the crystal, emerging on the other side as two mutually coherent beams. We have recently shown that a short acoustic pulse can coherently transfer energy between these two beams on a time scale consistent with the thermo-elastic model. Following the initial transient, the beam intensities oscillate as a function of the pump-probe delay. In the experiments reported here, the strain generation is studied as a function of the incident laser fluence. The relative phase of the oscillations and the amplitude of the transient provide information about the strain generation process at times shorter than the x-ray probe duration.

In the Laue geometry, two linearly independent wave solutions propagate through the crystal. Transverse to the propagation, these two solutions are standing waves whose wavelengths are twice the spacing of the diffracting planes. The solutions are usually labelled α and β with the convention that α has its nodes, and β its antinodes on the diffracting planes. In the case that all atoms lie on these planes, α is maximally transmitted and β is maximally absorbed. Because the two solutions interact with different electron densities, they propagate with different velocities.

Outside the crystal, two diffracted beams are produced: one in the direction of the input beam (forward-diffracted or “0” beam), and the other in the direction determined by the vector sum \( \vec{k}_H = \vec{k}_0 + \vec{G}_H \) (deflected-diffracted or “H” beam). Here \( \vec{k}_0 \) corresponds to the wavevector of the forward-diffracted (deflected-diffracted) beam and \( \vec{G}_H \) is the reciprocal lattice vector corresponding to the diffracting planes. These beams are linear combinations of the two internal solutions, α and β. The external intensities are given by:

\[
I_0 = |a E_0 e^{i\vec{k}_0 \cdot \vec{r}} + b E_\beta e^{i\vec{k}_\beta \cdot \vec{r}}|^2
\]  
\[
I_H = |c E_\alpha e^{i\vec{k}_\alpha \cdot \vec{r}} - d E_\beta e^{i\vec{k}_\beta \cdot \vec{r}}|^2
\]

where \( I_0(I_H) \) is the diffracted intensity of the forward (deflected) beam, \( E_{\alpha, \beta} \) is the complex wave field inside
the crystal, $\vec{k}_{\alpha,\beta}$ is the complex wavevector of the $\alpha, \beta$ solutions (including absorption), and $a, b, c, d$ are determined by the crystal orientation. The two internal modes $\vec{E}_{\alpha,\beta}$ oscillate in and out of phase as they propagate through the crystal. The wavelength of the interference, $\Lambda = |\vec{k}_\alpha - \vec{k}_\beta|^{-1}$, is known as the Pendellö sung length which is typically a few to tens of microns and is often shorter than the absorption length.

For a crystal that is thick compared to the $\beta$-absorption length, only the $\alpha$ solution survives and there are no interference effects. This is the anomalous transmission of x-rays, known as the Borrmann Effect [19]. A distortion of the lattice can cause a redistribution of the interior wave solutions [20]. Figure 1 shows the effect of a thin region of distortion regenerating the $\beta$ solution after it has decayed away in a thick crystal for the case of zero $\alpha$ absorption. When this occurs close enough to the crystal exit, the regenerated $\beta$ wave does not decay away and interference occurs at the exit face, despite the fact that the crystal is thick.

In our experiments, a short acoustic pulse is generated at the surface of a thick crystal. This pulse can be considered as a moving lattice disturbance. The diffracted intensities will oscillate in time as the pulse travels into the crystal bulk with a period that is given by the Pendellö sung length divided by the speed of sound. Deviations from the impulsive strain generation will be evident in the phase and/or amplitude of the x-ray modulation as a function of pump-probe delay.

The experiments were performed at the 7-ID undula-

tor beamline at the Advanced Photon Source. The x-ray energy was set to 10keV using a cryogenically cooled Si 111 double crystal monochromator leading to a $1.4 \times 10^{-4}$ fractional energy spread. The x-ray beam is masked by tantalum slits to ensure that the x-ray spot is smaller than the laser spot on the sample and to provide x-ray collimation. The sample is a 280$\mu$m thick, (001) Ge single crystal. The crystal was oriented such that the x-rays diffracted in the asymmetric 202 Laue geometry. In this geometry, and at 10 keV, the Pendellö sung length is 6.2 $\mu$m and the $\beta$ absorption length is 19$\mu$m, normal to the surface. Therefore, in the unperturbed crystal, only $\alpha$ survives at the exit. The only difference between the two diffracted beams is in their direction and a mismatch in their amplitudes due to details of the boundary conditions on the exit face [19].

Coherent strain pulses are produced on the x-ray output face of the crystal by sub-100fs, 800 nm laser pulses at a 1kHz repetition rate. The excitation is fully reversible between subsequent laser pulses. The laser is phase-locked to the x-ray beam to better than the x-ray pulse duration. The laser is timed to the x-rays using a combination of a digital delay generator and an electronic phase shifter in the phase-locked loop. In this manner the pump-probe delay may be set across a range of $\pm 1$ ns with 19 ps precision. A fast silicon avalanche photodiode (APD) and a picosecond x-ray streak camera [21] were used as the time-resolved detectors. The APD sampled the deflected-diffracted beam intensity and the streak camera sampled the forward-diffracted beam (see Fig. 2). The x-ray bunch separation was $\approx 152$ ns, large enough to allow electronic gating and measurement of a single x-ray pulse.

Following laser-excitation, high contrast oscillations are observed in the pump-probe data over a large span of excitation densities. Figure 3 shows these oscillations in the deflected-diffracted beam. The period of oscillation agrees with the Pendellö sung length divided by the longitudinal speed of sound. At an incident fluence of $35 \mu$J, the behavior near $t = 0$ shows a large transient that is unresolved with the 100 ps x-ray probe beam. After the transient, the oscillations show a significant phase-shift with respect to oscillations that occur following an excitation of $2 \mu$J. The amplitude and frequency of the oscillations are relatively insensitive to the fluence. However, as shown in Fig. 4, the phase is strongly dependent on the fluence and is correlated with the amplitude of the initial transient. The relative phase of the oscillation was defined with respect to the $2 \mu$J excitation and was retrieved from a least squares fit [22, 23]. The amplitude of the transient is defined as the diffracted intensity at a delay of 200 ps. Most of the energy transfer occurs in $\approx 40$ ps, measured with the forward diffracted beam using a streak camera (see the inset in Fig. 2). At relatively high fluences ($> 10 \mu$J), the intensity of the deflected diffracted beam approximately doubles while the forward
FIG. 3: Time-resolved anomalous transmission. The time-dependent intensity of the deflected-diffracted beam at three different incident optical fluences: 35 mJ/cm$^2$ (solid line), 7 mJ/cm$^2$ (dashed line), 2 mJ/cm$^2$ (dot-dashed line). Inset: Streak camera data showing the intensity of the forward-diffracted beam with picosecond resolution at an incident optical fluence of 35 mJ/cm$^2$.

FIG. 4: The relative phase of the Pendellöösung oscillations (squares) and the normalized deflected-diffracted intensity at a time delay of 200ps (circles) as a function of incident optical fluence.

diffracted beam is cut by more than 75%. At relatively low fluences (< 2 mJ/cm$^2$), there is no transient.

Inspection of (1) and (2) shows that the maximum energy transfer between the forward and deflected beams near the exit of a thick crystal occurs if the $\alpha$ and $\beta$ solutions are coupled at a depth of $\Lambda/4$. This implies that the transient behavior is due to a perturbation to the lattice that reaches a depth of more than 1.5 $\mu$m into the bulk. In the simplified picture that a moving interface couples the $\alpha$ and $\beta$ solutions, the excitation must propagate into the bulk at greater than 37,000 m/s, more than seven times the longitudinal speed of sound.$^{[24]}$

The strain pulse has a finite spatial extent and is comprised of a spectrum of phonons with different wavevectors. We expect that the phonon component with wavelength equal to the Pendellöösung length will resonantly couple the two interior wave solutions.$^{[23]}$ To model this phenomenon, we solve the equations of dynamical diffraction within the crystal, taking into account the laser-induced time-dependent strain profiles, using the Takagi-Taupin formalism adapted for Laue geometry.$^{[26, 27]}$ In this method, the differential equations coupling the $\alpha$ and $\beta$ branches are solved numerically. The depth-dependent strain profile for a given time is taken into account by noting that local strain is equivalent to a change in the local Bragg angle. Details of this approach (for Bragg geometry) can be found in the original work of Takagi$^{[26]}$ and Taupin$^{[27]}$. The means by which the method can be adapted for Laue geometry are implicit in the work of Zachariasen$^{[28]}$ and Batterman and Cole$^{[19]}$.

Pure thermoelastic models of strain propagation do not predict the observed fluence dependence of the phase and amplitude of the Pendellöösung oscillations. A proper model must include the effects of the coupling of the photoexcited plasma to the crystal lattice. The strain is comprised of both diffusive and elastic components: the diffusive strain is determined by the instantaneous temperature and carrier density profiles, modified by Auger recombination; the elastic strain is driven by changes in the temperature and carrier density, and propagates into the crystal at the speed of sound. In the absence of diffusion, a bipolar pulse develops in the time given by the optical penetration depth divided by the speed of sound.$^{[7]}$. For LA phonons with wavevectors along [100] and a 0.2 $\mu$m penetration depth, this corresponds to $\sim 40$ps. Including diffusion, the electron-hole plasma extends $\sim 1$ $\mu$m in the same time, leading to a strain front that has propagated into the bulk faster than the speed of sound.$^{[8]}$

Figure 5 shows the calculated diffraction intensity as a function of laser delay at an absorbed laser fluence of $3$ mJ/cm$^2$ (corresponding to a carrier density of $\sim 6 \cdot 10^{20}$ cm$^{-3}$). Good qualitative agreement with the experiment is seen (Fig. 3). The sharp initial rise in diffraction intensity is reproduced, as well as the frequency and phase of the time-resolved Pendellöösung oscillations. Figure 6 shows the calculated phase shift and the diffracted intensity as a function of absorbed optical fluence. After taking into account the surface reflectivity of the sample, good agreement with the experiment is obtained (Fig. 4).

In conclusion we have demonstrated a bulk sensitive probe of lattice dynamics using time-resolved x-ray anomalous transmission. We have observed that electron-phonon coupling modified by carrier diffusion is a dominant mechanism for energy transport in laser-excited Ge. This work could be extended to study how the elastic response of the material can modify the elec-
FIG. 5: Simulated deflected-diffracted intensity for an absorbed laser fluence of $3 \text{ mJ cm}^{-2}$.

FIG. 6: The calculated relative phase of the Pendellösung oscillations (squares) and the calculated normalized deflected-diffracted intensity at a time delay of 200 ps (circles) as a function of absorbed optical fluence.

Electronic transport properties of semiconductors.

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