Anomalous Elastic Response of Silicon to Uniaxial Shock Compression on Nanosecond Time Scales

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We have used x-ray diffraction with subnanosecond temporal resolution to measure the lattice parameters of orthogonal planes in shock compressed single crystals of silicon (Si) and copper (Cu). Despite uniaxial compression along the (400) direction of Si reducing the lattice spacing by nearly 11%, no observable changes occur in planes with normals orthogonal to the shock propagation direction. In contrast, shocked Cu shows prompt hydrostaticlike compression. These results are consistent with simple estimates of plastic strain rates based on dislocation velocity data.

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Although the response of materials to uniaxial shock compression has been a field of study for more than a century, our understanding at the lattice level of the response of crystals to rapid loading is still far from complete. While constitutive models are useful, a full description of phenomena such as shock-induced elastic-plastic flow and polymorphic phase transitions requires a knowledge of the atomic positions and the history of their rearrangement during the passage of the shock wave. In principle, one of the most direct methods of obtaining such information is the technique of *in situ* time-resolved x-ray diffraction (TXRD). Indeed, the TXRD experiments of Johnson and co-workers over three decades ago gave the first direct evidence of the retention of crystallinity under shock compression [1,2].

TXRD yields information about the interatomic spacings within the crystal. The change in Bragg angle due to the shock-induced alteration of the lattice parameter for monochromatic radiation is given, for small compressions, by simple differentiation of Bragg's law: $\Delta(2d_{hkl})/2d_{hkl} = -\cot\theta_b \Delta\theta$. TXRD also provides information about the degree of plastic flow within the crystal: in the limit of purely hydrostatic response, an initially cubic lattice remains cubic under shock compression (at least in the hard sphere approximation). Thus diffraction from planes with reciprocal lattice vectors orthogonal to the shock propagation direction also exhibits angular shifts: a feature that has been confirmed by TXRD for certain crystals such as LiF and KCl under sufficiently intense loading [3–5]. We provide here similar evidence for shocked single-crystal copper, which responds approximately hydrostatically to shocks of order 180 kbar on nanosecond time scales.

However, plastic flow relies on dislocation generation and transport, a process that takes a characteristic time τ_p. On extremely short compression time scales, τ_s, we might expect to observe purely elastic response within the crystal if $\tau_s \ll \tau_p$. An estimate of the time scale over which elastic response is maintained can be inferred from Orowan's equation [6]:

$$\frac{d\epsilon_p}{dt} = N|\mathbf{b}|v\,, \tag{1}$$

where ϵ_p is the plastic strain, N is the number density of mobile dislocations, $\mathbf{b}$ is the Burger's vector, and v is the component of average velocity of the dislocations parallel to the shock propagation direction. If the dislocation velocity is sufficiently low (as might be expected in covalently bonded materials), then even under large shear stress the time scale for plastic flow may be long.

In this Letter we present experimental time-resolved x-ray diffraction data that provide firm evidence that the response of single-crystal silicon to nanosecond time scale uniaxial shock compression along the (400) axis is anomalous in that it is purely elastic. Despite compressions along this axis of nearly 11%—more than twice the previously accepted steady-state Hugoniot elastic limit [7]—the (040) diffraction data, which measure the unit cell spacing orthogonal to the shock propagation direction, confirm the lack of detectable atomic motion perpendicular to the shock front. For similar shock strengths, however, single-crystal Cu, shocked along the (200) axis, exhibits compression of the unit cells along both (200) and (020), indicating a prompt transition to plastic flow.

The angular width of the Si (040) peak is sufficient to confirm the low density of dislocations or stacking faults within the shocked region. Although little is known directly concerning dislocation velocities under the ultrahigh shear stresses present within the shocked crystals, extrapolation of dislocation velocities calculated for silicon at lower shear stresses indicate that plastic flow may take of order milliseconds, at least for the dislocation densities inferred here, and would still be of order microseconds at the limits of possible dislocation densities. The results indicate that we can study, at the lattice level, a novel regime of matter where crystals are compressed in one dimension without plastic flow.

The experiment with Si, shown in schematic form in Fig. 1(b), was performed using the Nova laser at Lawrence Livermore National Laboratory [8]. The laser provided both the pressure source with which to shock compress the single-crystal silicon (via a laser-heated hohlraum) and the quasimonochromatic x-ray source for subsequent TXRD. The target consists of a millimeter scale cylindrical gold cavity (hohlraum) with internal shielding [9]. The thin silicon crystal is mounted over a hole in the central cavity of the hohlraum target, and a Fe foil, from which the x-ray source is generated, is positioned off to the side. Nova is a large laser with ten beams that enter the target chamber in two opposing cones, each with a 50° half angle. Eight beams enter the target through the laser entrance holes on each end.

Energies of 2.4, 3.3, and 10.5 kJ of 0.35 μm light are delivered to the target in a 4 ns square pulse for low, intermediate, and high drive cases, respectively. The laser light heats and ionizes the inner surface of the gold wall in the two laser-heated cavities to create a high-temperature plasma. Gold M-band (2–3 keV) and thermal extreme ultraviolet (XUV) radiation generated from the plasmas heat the central cavity by passing through holes in the internal shields. A nearly Planckian XUV field is created in this central cavity, with a radiation temperature increasing from 20 eV to 40–60 eV over a 3–4 ns interval. The internal shielding is positioned within the hohlraum so that there is no direct line of sight to the crystal from the laser plasmas where the M-band x rays are generated, preventing the Au M-band emission from the laser-heated spots preheating the crystal prior to shock compression.

The quasiblackbody XUV radiation ablatively drives a shock into the 40-μm-thick, 0.8 $\times$ 2.0 mm, silicon single crystal which is mounted over a hole in the side of the hohlraum at the midplane with the (400) planes parallel to the surface of the crystal to within 0.5°. Two additional beams of Nova generate the x-ray source ("backlighter") used for Bragg diffraction from the (400) planes. These beams are focused onto a separate 5-μm-thick Fe foil located behind a Ta pinhole aperture. This provides a 150 μm diameter x-ray source. Heliumlike ionized Fe emits K-shell line radiation, predominantly in the 6.7 keV resonance line at a wavelength of 1.85 Å. The Fe foil is located approximately 2 mm from the center of the crystal. The crystal subtends an angle of about 25° to the x-ray source. The $2d$ spacing of the unshocked (400) lattice planes is 2.71 Å giving a Bragg diffraction angle for the unshocked Si of 42.95°. The x rays Bragg diffracted from the (400) planes of the crystal are recorded with an x-ray streak camera which has a 3-cm-long photocathode that is located 123 mm from the target, allowing us to collect diffracted x rays over a range of Bragg angles of nearly 15° with better than 100 ps temporal resolution. A film holder is placed in front of the streak camera to record simultaneously a time-integrated image of the diffracted x rays, as shown in Fig. 1(c). In addition, x rays diffracted from the atomic planes with reciprocal lattice vectors orthogonal to the shock propagation direction—the (040) planes—and transmitted through the crystal were recorded simultaneously [see Fig. 1(a)]. Such diffraction through a crystal is often termed Laue diffraction, and gives us direct information about the deformation of the crystal perpendicular to the shock propagation direction.

For the majority of data shots the Laue signal was recorded on static film, but for a small number of shots the target configuration was reversed to record the time-resolved Laue (040) signal. The time-resolved record for the (400) Bragg diffraction data shown in Fig. 1(c) is given in Fig. 2. For this shot the ("drive") laser energy incident in the hohlraum was 2.4 kJ, and the 4 nsec pulse used to

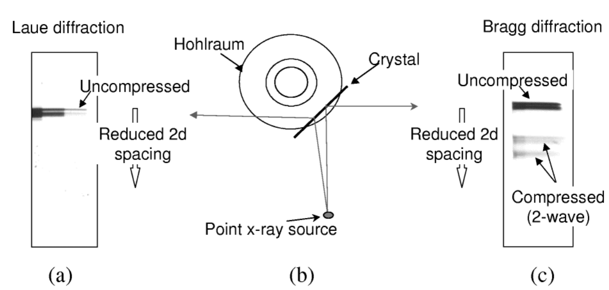

FIG. 1. A schematic diagram of the experimental setup is shown in (b), along with time integrated (a) Laue and (c) Bragg x-ray diffraction signals (see text for details).

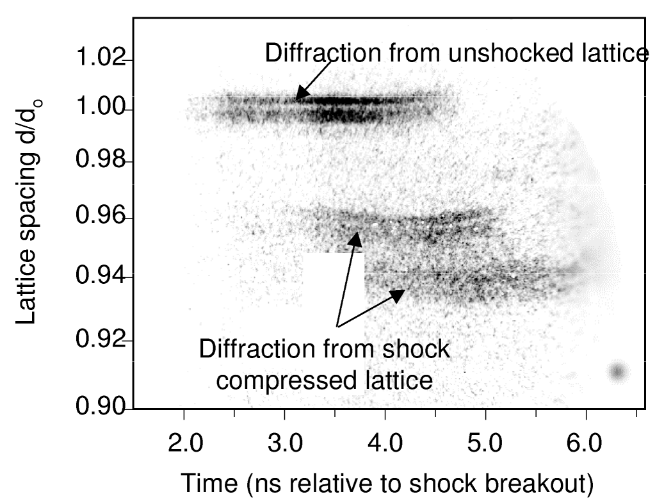

FIG. 2. X-ray streak camera record of the Bragg signal shown in Fig. 1(c). The diffraction angle and corresponding compression of the (400) lattice are shown as a function of time with respect to the start of the drive laser.

produce the backlighter x-ray diffraction source was timed starting at 2 nsec after the drive laser turned on. Note from Fig. 2 that x rays are Bragg diffracted from both uncompressed and shock compressed (400) lattice planes. The mean-free path length of 6.7 keV x rays in Si is about 40 μm. Hence, we volumetrically probe a substantial portion of the sample and simultaneously observe regions of both shock compressed and uncompressed Si crystals, as the shocks move through the target. From Fig. 2 it can be seen that for this particular shot the x rays sampled waves with two distinct lattice spacings, corresponding to compressions of the (400) planes of 4.3% and 6.2%. The two distinct spacings can be seen even more clearly on the corresponding time-integrated signal in Fig. 1(c). Profiles of the time-integrated data shown in Figs. 1(a) and 1(c) are given in the upper traces of Figs. 3(a) and 3(b). Remarkably the diffraction from each of the two distinct regions of compression clearly shows the two main x ray lines from the Fe heliumlike ion at 1.850 and 1.868 Å, [see also Figs. 1(c) and 2], indicating a low strain gradient (i.e., plateau) in these compressed regions. Indeed, it is possible to extract strain-depth information from the data: simulated diffraction, assuming a given strain-depth profile, can be generated from computed solutions of the Takagi-Taupin equations [10–12] and then an iterative procedure used to extract strain-depth profiles, as has been used previously to determine strain profiles in laser-irradiated semiconductors [13]. This procedure indicates that the initial wave has an extremely steep rise from 0% to 4.3% compression in less than 1.5 μm: a strain rate greater than 2×10^8 s^{-1}. The faint diffracted x rays between the two regions of distinct compression are consistent with an approximately linear rise in compression from 4.3% to greater than 6% over a distance of $\sim$5 μm. Also shown in Figs. 3(a) and 3(b) are lineouts from higher drive energy shots, with the highest compressions in the Bragg direction of nearly 11% being recorded. Note that, even for these higher compression shocks, there is no noticeable change in the Laue signal, indicating that the crystal response is purely elastic.

A two-wave shock structure in some of the data shots implies regions with different compressions due to shocks of different strengths. This is most likely caused by the increasing ablation pressure due to the hohlraum radiation temperature, which increases from 20 to 40–60 eV over a 3–4 ns interval, leading to compression profiles which are sensitive to the drive history. Simulations (treating the Si elastically) show that a first shock is launched early in the drive, and that two, and perhaps three, compression waves can be launched as the drive increases in strength. The radiation temperatures are calculated from 2D hohlraum simulations, using the time-dependent laser powers from each shot. This leads to drive temperatures that increase from 20 eV early in time to peak values of 40, 45, and 61 eV at 4 ns for the low, intermediate, and high drives, respectively. The longitudinal component of the stress tensor for these elastic simulations at 4.5 ns (which is just prior to shock breakout from the back of the Si foil) has leading edge and peak values of 80 and 190 kbar (low drive), 120 and 260 kbar (intermediate drive), and 240 and 600 kbar (high drive).

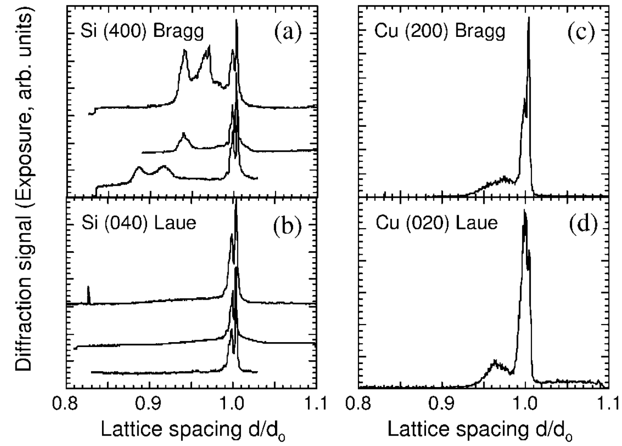

FIG. 3. Profiles of the time-integrated x-ray diffraction signals from laser-shocked single-crystal (a) Si in the Bragg (400) geometry, (b) Si in Laue (040) geometry (upper traces correspond to the shot shown in Figs. 1 and 2, with a peak compression of 6.2%, the middle traces for a shot with similar peak compression, but slightly different drive history, and the lower traces for a shot with peak compression of 11%), (c) Cu in the Bragg (200) geometry, and (d) Cu in the Laue (020) geometry.

While compression is clearly seen within all the Bragg signals, the time-integrated image of the Laue (040) diffraction [Fig. 3(b)] shows no compression, broadening, or shift caused by the passage of the shock wave. Indeed, the Laue data show a spectrum of the heliumlike Fe radiation as well resolved as a perfect crystal. For those shots where this signal was time resolved, no alteration in the diffracted intensity was observed, even for times corresponding to the whole of the crystal being traversed by the shock wave. The x-ray structure factor (and thus intensity) for the (040) reflection is simply a function of the atomic positions orthogonal to the shock propagation direction, and thus this data constitutes clear evidence for a lack of atomic motion in the Laue planes. Furthermore, the absence of observable broadening allows us to estimate an upper limit to the number of dislocations present within the crystal. The spectral width of the heliumlike Fe lines corresponds to an angular width of 0.2°: the lack of broadening of the Laue signal with this angular resolution implies that the density of dislocations or stacking faults was significantly below 10^{10} cm^{-2}. Similar results have been obtained with simultaneous TXRD and velocity interferometry diagnostics using the TRIDENT laser at Los Alamos National Laboratory, where a two-wave structure was evident in the Bragg signal and the velocity history of the free surface, but the Laue signal was not affected by the shock [14]. The effects noted above have been seen over a wide range of compressions, up to a peak compression of the (400) planes of 11% (limited by the shock pressure we could generate using shielded hohlraums of the dimensions cited above).

These results strongly suggest that the response of the silicon is purely elastic. *Ab initio* quantum mechanical simulations of uniaxial compression of Si predict that the shear stress does not increase monotonically with strain, but exhibits structure which may limit the generation and motion of dislocations [15].

While such purely elastic response might at first seem remarkable, we note that dislocation velocities within Si are known to be slow, as is typical for covalently bonded materials [16,17]. While neither experimental data nor calculations exist for the high stresses implied here (of order 100–600 kbar), we note that several calculations have been performed at stresses of order 1 kbar [18]. Linear extrapolation of this data to the stresses encountered here implies dislocation velocities of order 0.1 $\mathrm{mm\,s^{-1}}$. From Orowan's equation [see Eq. (1)], using $b = 3.83$ Å, $v = 0.1\ \mathrm{mm\,s^{-1}}$ [19], and the upper bound on the dislocation density of $10^{10}\ \mathrm{cm^{-2}}$ inferred from the diffraction signal, we deduce that it would take longer than 10 ms to achieve a plastic strain of 5%. Clearly, higher dislocation densities, which may be generated on longer time scales, would reduce this figure, but even dislocation separations on the scale of a Burger's vector would only allow plasticity on a 10 ns time scale—significantly longer than those probed here.

Clearly, in materials where the dislocation velocity is high, such elastic effects will only occur on very short time scales. For example, the dislocation velocity in Cu is known to be approximately 6–7 orders of magnitude greater than that in Si [19]. Thus we might expect that plasticity can occur on subnanosecond time scales for Cu. We have performed experiments on 2-μm-thick single-crystal Cu similar in setup to that described above for Si. The experiments were performed on the Omega laser at the Laboratory for Laser Energetics at the University of Rochester [20], and only differed from the Si work in that the shock pulse was provide by direct illumination of the crystal with the laser beam, rather than via the XUV radiation within a hohlraum. The direct-drive laser pulse corresponded to an average intensity of $4 \times 10^{11}\ \mathrm{W\,cm^{-2}}$ over 3.5 ns in a 2.5×2.7 mm spot, using 70 J at 0.35 μm wavelength. The pulse shape had a 2 ns linear ramp, followed by a 1.5 ns constant intensity interval. Profiles of the time-integrated images of the diffraction of a 4 ns pulse of 2.38 Å radiation from the (200) and (020) planes are shown in Figs. 3(c) and 3(d). The 4 ns x-ray pulse diffracts from the crystal both before and after the onset of shock compression, and thus the position of the diffraction from both the undisturbed and shocked lattice can be seen. In this case, in contrast to the silicon data, the cubic lattice has been compressed in all dimensions, indicative of plastic flow. The volume reduction is of order 8%, corresponding to a hydrostatic pressure of order 180 kbar. Note that for Cu the diffracted signal from the strained regions is broad, consistent with a high fault density. Applying Orowan's equation in this case, using $N = 10^{11}$–$10^{12}\ \mathrm{cm^{-2}}$ (consistent with the width of the diffraction peaks), $b = 2.56$ Å, and $v = 10^6\ \mathrm{mm\,s^{-1}}$ [21], gives a characteristic time of 10–100 ps for the transition to plastic flow, consistent with our observations.

In conclusion we have employed *in situ* TXRD with subnanosecond temporal resolution to measure the lattice parameters of orthogonal planes in shock compressed silicon and copper single crystals. Despite uniaxial compression of the (400) planes of Si by nearly 11% for nanosecond time scale compressions, we find no change in the (040) lattice parameter, indicating that even under such high compressions the response of Si is purely elastic. The results are consistent with the extremely low dislocation velocities expected for silicon, and are in marked contrast to experiments on Cu, where higher dislocation velocities are expected and hydrostatic compression observed. These results demonstrate the capability of studying the onset of plastic flow at the atomic level.

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