Orthogonal strains and onset of plasticity in shocked LiF crystals

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We report experimental determinations of the elastic component of the transverse strain during passage of a shockwave. A 1 ns laser pulse drove an ≈ 60 kbar shock into an aluminum coated, radiation hardened, pure LiF single crystal. By diffracting a 0.25 ns pulse of x rays from a nearby high brightness, time-synchronized laser plasma, we measured the simultaneous compression of the lattice both parallel and transverse to the shock propagation direction with subnanosecond resolution. Definite compression was recorded in the inertially confined transverse direction, indicating the onset of plastic lattice response. Peak strains were of comparable magnitude in the orthogonal directions, but were not equal.

Pulsed x-ray-diffraction experiments in the 0.13–1.1 Mbar range were performed in the 1970s on shocked LiF with tens of ns resolution and using explosive[1] or gas gun drivers.[1,2] Those experiments established the conclusion that compression of single crystals by shock waves (uniaxial) is hydrostatic (isotropic) and therefore plastic on those time scales, but in a manner which somehow preserves single-crystal orientation. These findings and their juxtapositions of symmetries and orientations contrast with experimental measurements[3] at a few tens of kbar, which support the view[4] that the immediate and momentary response of LiF single crystals to flyer plate impacts is elastic and uniaxial in strain. The relaxation of impact stress in LiF has been measured[5] by many workers out to several microseconds during plate impacts ≤ 30 kbar on soft, thermally annealed single crystals with known levels of impurities. The phonon-mediated relaxation of stress in perfect Leonard-Jones crystals has been simulated by molecular dynamics, with discernible transverse effects coming to rapid completion within a few tens of atomic layers of shock transit.[6] In previous work it has not been possible to independently determine a transverse component of either stress or strain directly. It is therefore of interest to make observations, during a shock, of directional lattice strains, on a nanosecond time scale.

We report simultaneous measurements of the elastic component of strain in axial (parallel to shock propagation direction) and transverse (perpendicular) directions during shock passage through a LiF single crystal. At 1.0 ns after the peak of the 1.1-ns shock driving laser pulse, the crystal has achieved a transverse peak compression of magnitude comparable to the axial peak compression, as expected near hydrostatic conditions. Shock-induced intensity and angular variations in the transverse diffraction angle imply the onset of plasticity in the laterally inertially confined lattice.

The experimental arrangement (Fig. 1) differs from that in previous papers[7–10] by the addition of a (002) diffracting beam and film pack. LiF single crystals, radiation-hardened UV grade, were cleaved as received from Harshaw, thinned and polished to grit size 1 μm, and coated with 1000-Å Al and 25-μm $(CH)_n$ parylene. Typical finished sizes were 7 mm$\times$15 mm$\times$0.08 mm. The CH was semitranslucent, which inhibits visible interference fringes and may enhance irradiation uniformity on the Al. Laser pulse widths were measured by streak camera. Error bars on all reported measurements are typically within $\pm 10\%$. A single, 1.1-ns FWHM, $\lambda = 1.06$-μm beam of the JANUS Research Laser at the Lawrence Livermore National Laboratory, focused to 5×10^9 W/cm^2 over a diameter of ~ 15 mm, ablated the Al to drive a shock wave axially into the LiF in the **x** or [100] direction. The CH lengthens and strengthens the pressure pulse to about 60 kbar, comparable to our previous experiments.[9] (Numerous plate impact experiments have shown that the dynamic yield point or HEL of LiF is not a single number, but varies with sample preparation and shows stress relaxation over a range 2–25 kbar.) A driver beam block (x-ray transmissive 6-μm Mylar with an ~ 1000-Å Al flash coating) shadows half of the 7 mm dimension of the target and provides a sharp demar-

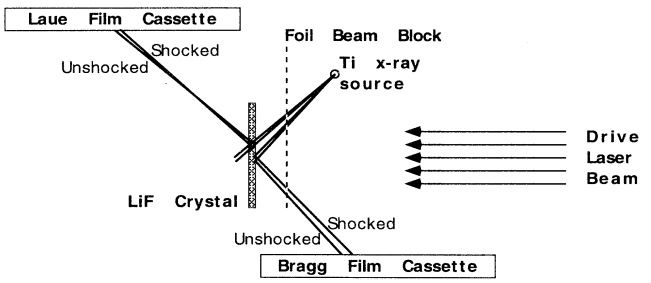

FIG. 1. Setup showing 1.1-ns shock-driving IR laser beam, crystal target, Ti x-ray source at focus of 0.2-ns green laser beam, and Bragg and transmission x-ray diffraction paths to two x-ray film packs. Incoming IR beam is blocked (dotted line) from irradiating the left half of the crystal (out of the plane of the page). Cassettes do not view the source.

cation between shocked and unshocked crystal. A second, $\lambda=0.53$ μm, 0.25-ns laser beam was tightly focused ($\sim$50-μm spot) onto a nearby Ti target at a delay time of 1.0 ns after the peak of the 1.06-μm pulse. Pulsed, monochromatic, spherically divergent x-ray lines from highly ionized Ti diffract from the LiF crystal at Bragg angles determined by the strain-dependent lattice spacing $2d$. We shall refer to diffraction from (200) planes, parallel to the shock front (unshocked $2d=4.028$ Å), as Bragg diffraction, and that from the orthogonal (002) planes, requiring x-ray transmission through the entire crystal thickness, as Laue or transmission diffraction. The Bragg film records axial compression, and the Laue film is sensitive to transverse strains. By symmetry, the (020) planes are redundant to the (002) planes in this experiment, and therefore were not monitored. Kodak DEF x-ray films in separate cassettes recorded the diffraction patterns. Each shot required a new crystal target. Several shots suffered from unacceptably long (>0.3 ns) x-ray pulses and were not included in the final data. Of the five remaining shots taken with the above parameters, three provided sufficiently complete data to analyze quantitatively and report in this paper. All shots, even those not reported, exhibit the same qualitative features described below.

The rise times of our pressure pulses are within the 1–2 ns rise times reported as sharp shocks by Asay *et al.*[11] in studies of elastic precursors in LiF. The total elapsed time allows for only a few tens μm travel in any direction for shock or release waves, thus edge effects from lateral sample size are precluded, and the lattice is constrained in the transverse direction by inertial confinement. For a laterally uniform laser drive, the shock wave will be uniaxial in strain, although strain will vary with depth and time due to the laser's Gaussian temporal profile. Elastic strains are compatible with diffraction because crystal order is retained. Plasticity is inherently a disordering of the lattice, and is determined by inference from the elastic diffraction signal. The diffraction signals were examined for intensity changes and for angular changes.

The diffraction films for (200) and (002) planes are reproduced in Figs. 2(a) and 2(b), respectively. The film images show uniform intensity along the lines in the unshocked region (occasional irregularities are from imperfections in the foil beam block). The source spectrum, best resolved in the unshocked traces, consists of one group of closely spaced lines[12] near 2.63 Å, sufficiently intense to probe the shocked crystals, and higher Rydberg series members (not shown), useful for wavelength calibration.[13] On the high-angle, long-wavelength side of the well-defined lines lies a region of exposure by blended lines diffracted from the transiently compressed lattice planes, the profile of which is determined by the strain as a function of depth. The smooth texture (disregarding film grain) of the shocked signature indicates a comparatively uniform laser irradiance. The loci of diffraction for the uncompressed (200) and (002) planes approach each other within $\sim$2 mm on the crystal surface, the shocked loci being a bit closer together.

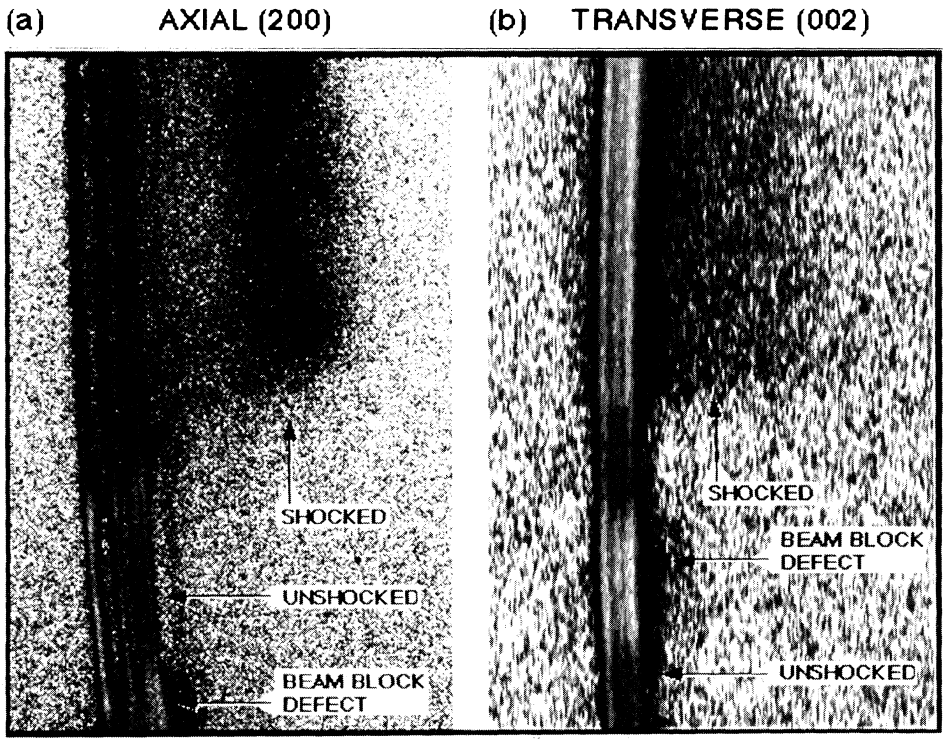

FIG. 2. Diffraction films for 1.1-ns delay (shot 25-2): (a) axial; (b) transverse, entire image processed to enhance visibility of shocked signature.

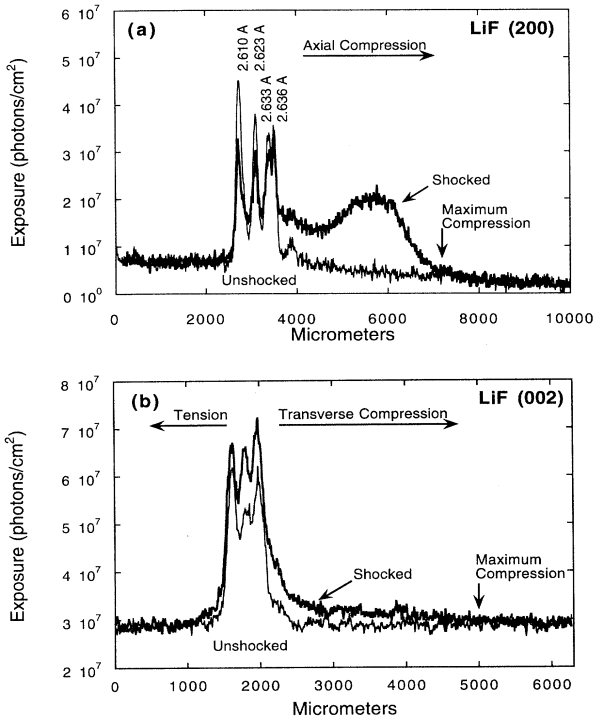

FIG. 3. Profiles of films of Fig. 2: (a) axial; (b) transverse.

The profiles of the intense Bragg lines are shown in Fig. 3(a). The exposure level of the lines from the unshocked region is only slightly reduced (6%) by x-ray absorption in the laser beam block. The shocked signature is well removed from the original lines. The integrated exposure of the shocked signature is significantly greater than that of the unshocked crystal. Theoretical calculations of dynamical diffraction in 0.08-mm-thick LiF at 2.63 Å give a single crystal $R=4.1\times10^{-5}$ for a perfectly ordered lattice, and up to $R=5.0\times10^{-4}$ for the kinematic "ideally imperfect" Bragg case. In laboratory LiF crystals, where enhanced reflectivity correlates directly with defect density and rocking curve broadening, kinematic values are not achieved.[14]

However, transiently shocked samples may surpass quiescent reflectivities, i.e., have higher defect densities or elastic strain gradients and therefore have higher reflectivities more closely represented by the ideally imperfect theoretical case. These increases in reflectivity are related to x-ray probe depths. The Bragg probe depth in perfect unshocked crystal is set by primary extinction,[16] by which diffraction from the perfect lattice rapidly deflects the rays out of the crystal and limits penetration depths to only 4 μm below the surface ($\mu_c=2.5\times10^3$ cm^{-1}). In shocked crystal, the compression gradient circumvents primary extinction by altering the lattice spacing, permitting the x rays to penetrate more deeply. The ultimate Bragg probe depth in shocked material is set by photoelectric absorption, which suffers a $1/e$ falloff over a ray path of ~70 μm (Bragg depth 23 μm). Thus, the shocked crystal can involve a much larger volume of crystal in the diffraction process and so enhance the diffracted power. The integrated intensity in the shocked region exceeds that in the unshocked region due to this kinematic enhancement,[15] but the undeflected intensity in the shocked region drops, due to diffraction and absorption in the overlying thickness of shocked crystal.

Measurement of the maximum $d\vartheta$ in the shocked signature (Fig. 3) leads to the peak compression through the differentiated Bragg law, $dd/d\vartheta=-d\cot\vartheta$, where ϑ is known for uncompressed value $2d$. The measured axial values of peak strain dd/d were -5.0, -5.2, and -5.1 %, which represents the peak strain of that part of the crystal where compression is sufficiently elastic to support diffraction. These values are consistent with the irradiance (drive pressure), the delay time (temporal profile of pressure),[9] and the LiF Hugoniot.[17]

The transmission film profile [Fig. 3(a)] is quite different than the Bragg [Fig. 3(b)]. The Laue undeflected signature actually increases in the shocked region, contrary to the Bragg result (above) for the same region. X-ray absorption in the beam block, a mere 3%, cannot account for the increase.[15] The enhancement can only arise from an increase in diffracting volume of crystal. In the Laue geometry, the volume cannot be increased by the probe depth, since all rays must transit the entire crystal and probe depth is a constant. The additional volume is achieved laterally by a strain-induced lowering of lattice perfection, as discussed above but now applied in the transverse direction. As a result, reflectivity in the shocked crystal is kinematically enhanced, and the shocked diffraction is added to that from the as yet unshocked crystal, even though unaccompanied by a discernible change in angle.

The exposure of the deflected shock signature on the Laue film is reduced by absorption in the entire crystal thickness, and is lower than that of the Bragg film. The compressed Laue signature shows a gradual decrease in exposure from the original line position out to the maximum angular displacement for the shock. In the transmission direction, the maximum deflection yields peak strains for the three shots of $dd/d=-6.6$, -7.8, and -10.0 %, an average of $-8.4(\pm1.4)$%. The reason for the variability in the Laue direction is unexplained, but may be partially due to the ≈0.1 ns uncertainty in laser timing. The peak strains in the Laue direction, measured at 1.0-ns delay time, are comparable to the expected axial strains at the temporal peak of the pulse.

Ascribing the average peak elastic strains in orthogonal directions to the same local volume of crystal corresponds to compressed unit-cell dimensions of 3.82 Å axially and 3.69 Å transversely, compared to the uncompressed $a=4.028$ Å.

In the transverse direction, where inertial confinement leads to the constraint of zero net strain,[18]

$$\epsilon_e+\epsilon_p=\epsilon_T=0\ , \tag{1}$$

where the total strain ϵ_T is zero, so that the plastic strain ϵ_p (due to disorder in the lattice) is deduced as the negative of the elastic strain ϵ_e measured by diffraction from the remaining ordered lattice. The above measurement of dd therefore quantifies the transverse plastic strain.

Stated another way, the elastic strains shown by the Laue films are direct evidence that compensating plastic strains have been produced according to the requirements of lateral inertial confinement. Both the intensity and angular data corroborate the existence of these strains, and are consistent with this finding.

The discrepancy between strains in the orthogonal directions ($dd/d = -5.1\%$ axially, and -8.4% transverse) may be related to the fact that we are observing the strains after the peak of the laser pulse. Earlier measurements on Si targets under comparable conditions demonstrated a relaxation of axial compression at this delay time, due to a drop in laser irradiance.[9] Elastic strain relaxation in the axial direction is unconstrained and may proceed immediately, analogously to the rapid initial rise of elastic strain.[4] In the transverse direction, however, the total strain remains inertially confined and subject to Eq. (1). Our observation of heightened transverse strain implies that relaxation is less complete transversely than axially, and that the time scale of the relaxation is accessible to measurement. This time scale is intermediate between the femtosecond time scale of molecular-dynamics simulations[6] and the tens of nanoseconds found experimentally in shock-induced stress relaxation and phase transitions[19] in alkali halides.

A small tensile shift is seen in the transverse direction (a yet lesser tensile signature may also be detectable in the Bragg traces). The maximum angular displacement corresponds to $d\vartheta = 0.016$ rad, or a 2.9% transverse extension in d within some small fraction of the volume. Thermal expansion could be responsible, but is not expected on these time scales and was not observed in earlier related experiments on Si.[10] The present data are insufficient to determine whether the tensile contribution is a portion of a profile broadened by dislocations and plastic flow, or whether simple tilting of compressed planes is producing a shift. Tilting is known to have occurred at late times in impactor tests.[2] If tilting is present here, then a very high angular velocity is being produced, so high that particles larger than a critical size would exceed the material strength. In other shots taken during this experiment, we observed structures of size consistent with this critical size, and which become increasingly more well defined at later delays.[7]

In summary, we have observed the onset of plasticity in a single crystal subject to shock, have measured local, directional elastic strains in the principal directions of the unit cell, and have inferred plastic strains in the inertially confined transverse direction. Peak compressions approach hydrostatic conditions, although deviations from hydrostaticity are observable. The results exemplify the unique capabilities and excellent temporal resolution of the pulsed x-ray technique and call for improvements in laser illumination of large areas (several cm^2) with longer duration, more sharply rising pulses at constant but moderate irradiances.

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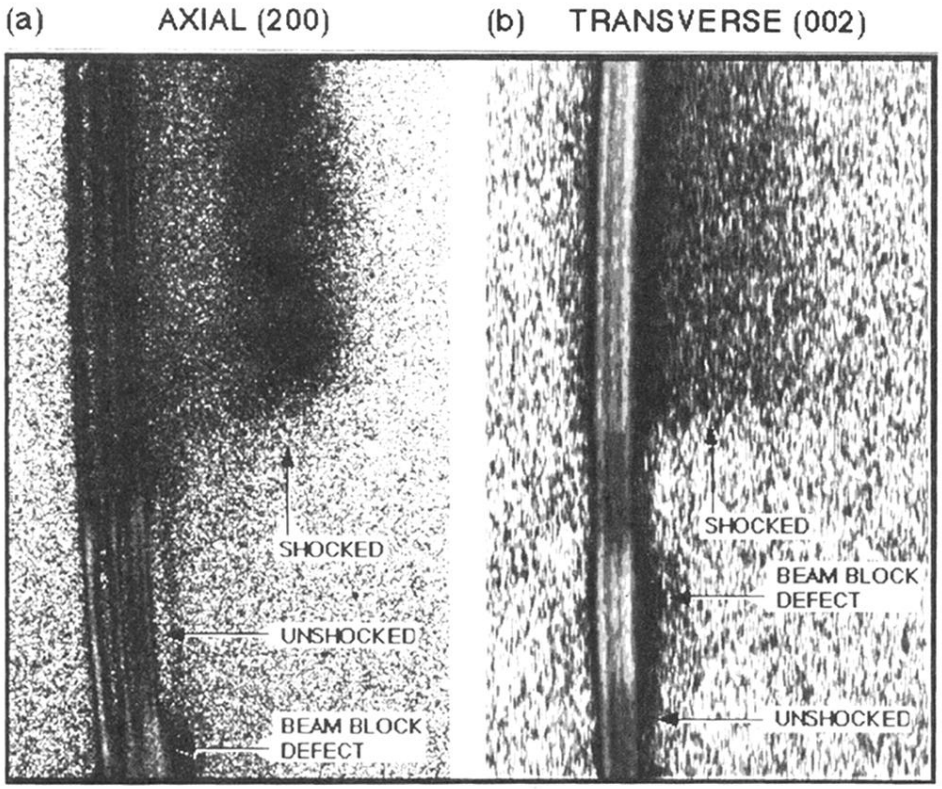

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