

Investigations into rapid uniaxial compression of polycrystalline targets using femtosecond X-ray diffraction

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Abstract. Although the pressures achievable in laser experiments continue to increase, the mechanisms underlying how solids deform at high strain rates are still not well understood. In particular, at higher pressures, the assumption that the difference between the longitudinal and transverse strains in a sample remains small becomes increasingly invalid. In recent years, there has been an increasing interest in simulating compression experiments on a granular level. *In situ* X-ray diffraction, where a target is probed with X-rays while a shock is propagating through it, is an excellent tool to test these simulations. We present data from the first long-pulse laser experiment at the MEC instrument of LCLS, the world's first hard X-ray Free Electron Laser, demonstrating large strain anisotropies. From this we infer shear stresses in polycrystalline copper of up to 1.75 GPa at a shock pressure of 32 GPa.

1. Introduction

Despite many decades of study, how materials respond at ultra-high strain rates (10^6 – 10^{10} s⁻¹) is not well understood. Of particular interest is how solids resist deformation by supporting shear stresses, while they transition from elastic to plastic behaviour. The shear stress supported is a measure of material strength, which is arguably the most important parameter defining plastic flow and, as such, a number of theoretical strength models have been developed to understand plasticity [1–4]. There is a large array of studies into the material strength of samples that have been shocked by either explosive lenses [5–7] or gas guns [5, 8–11], with the different stress components being measured by stress gauges. These techniques are limited both in terms of the pressures at which measurements can be made (tens of GPa) and the strain rates achievable ($\sim 10^6$ s⁻¹). However, by performing *in situ* X-ray diffraction on a laser-shocked target, measurements of the anisotropy between transverse and longitudinal strains have been made up to 100 GPa in single crystals [12]. This work then inferred stresses from the measured strains using MD simulations, in order to determine the material strength, as exhibited by the



residual shear stress. Later, results were obtained in polycrystalline iron targets by Hawreliak *et al.* [13], in which a different, tilted target geometry was used. By breaking the symmetry of the experiment, Hawreliak was able to measure different components of the strain tensor by observing the variation in the implied interatomic planar spacing around the Debye-Scherrer ring. However, no material strength was seen to within experimental error, perhaps owing to the stress relief due to the $\alpha - \epsilon$ phase transition in the Fe. The work presented here builds on this technique, replacing the laser-plasma X-ray backlighter with the beam from an X-ray Free Electron Laser (FEL) as the X-ray source, offering a number of advantages. With a temporal resolution of ~ 100 fs, the Linac Coherent Light Source (LCLS) can effectively freeze atomic motion, since the pulse length is comparable to the shortest phonon period. Furthermore, the beam is bright enough to capture a complete diffraction pattern in just a single shot. Lastly, LCLS offers an order of magnitude improvement in bandwidth compared to existing laser plasma sources, providing the resolution to measure much smaller strain anisotropies.

2. Experimental Details

An experiment was performed using the MEC (Matter in Extreme Conditions) instrument of LCLS at SLAC National Accelerator Laboratory. A $20\ \mu\text{m}$ thick polycrystalline Cu foil obtained from Goodfellow, was shocked with a 10 ns, 527 nm square laser pulse of differing energy focused to spot sizes of either $250\ \mu\text{m}$ and $200\ \mu\text{m}$, giving irradiances of $2.7 \times 10^{12}\ \text{Wcm}^{-2}$ and $1.6 \times 10^{12}\ \text{Wcm}^{-2}$, at 35° and 25° to the target normal respectively, resulting in induced pressures of a few tens of GPa. Furthermore, since the diameter of the FEL beam was reduced to $50\ \mu\text{m}$, much smaller than the spot size of the drive laser, we can assume that an even pressure is applied to the region of interest by the drive laser.

After a variable time delay, the LCLS beam at a photon energy of 8.2 keV was used to interrogate the sample at 45° to the sample normal, producing a Debye-Scherrer diffraction pattern. By having the target normal at an angle to the X-ray beam, the symmetry of the system was broken and the angle between the reciprocal lattice vector, \mathbf{G} , and the target normal, ψ , now varies as a function of azimuthal angle, ϕ (see figure 1). Thus, by observing the diffraction

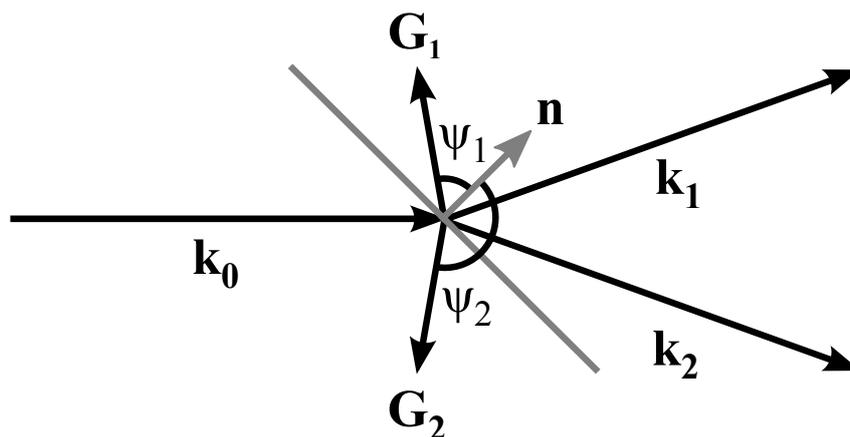


Figure 1. By tilting the target relative to the direction of the incoming X-rays, the angle between the reciprocal lattice vector, \mathbf{G} , and the target normal, ψ , changes as a function of the azimuthal angle, ϕ .

pattern at different points of the Debye-Scherrer ring, information can be gained about grains in the sample with different \mathbf{G} vectors. Two Cornell Stanford Pixel Array Detectors (CSPADs) [14] were placed around the sample in order to probe a number of diffraction lines from both driven and undriven parts of the sample (see figure 2).

The top transmission CSPad (D1) was almost completely sensitive to only transverse strain, whereas the top reflection CSPad (D2) was sensitive to both longitudinal and transverse strain.

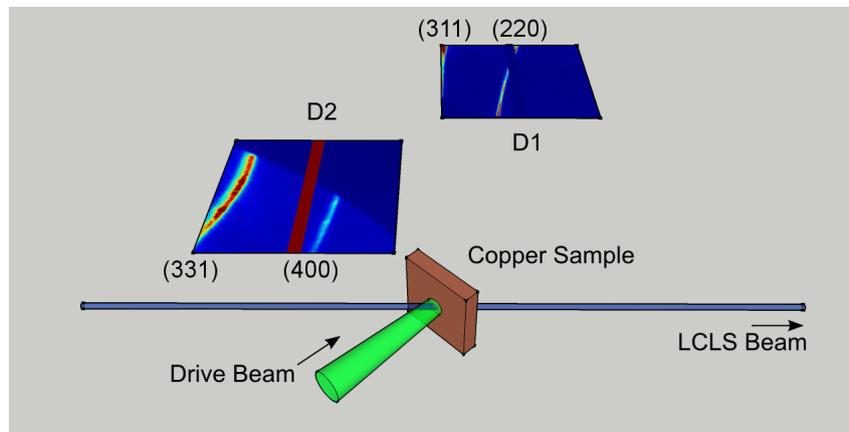


Figure 2. A schematic diagram of the experimental setup. The copper polycrystalline target is tilted at 45° to the LCLS beam. Two CSPads were placed to record different parts of the diffraction pattern.

3. Analysis

Analysis of the data requires an accurate knowledge of the position and orientation of the two detectors with respect to both the shocked target and the LCLS X-ray beam. Although these positions were recorded mechanically with reasonable accuracy during the experiment, it was found that more exact relative positions could be found from fits to the registered Debye-Scherrer rings from unshocked samples. Images of the diffraction pattern from the undriven targets were captured, and the background counts subtracted. The positions of the different Bragg peaks were used in a constrained fitting algorithm, which then found the positions and orientations of each of the detectors.

Whilst analysing data from driven samples, it should be noted that it is not possible to find components of the strain tensor by simply dividing the interatomic planar spacing obtained from driven and undriven samples using Bragg's law. This is because in the large strain limit, the diffraction from the driven and undriven data will originate from different sets of diffracting grains. For any general diffracting plane in a polycrystal, the outgoing \mathbf{k} vector will be given by $\mathbf{k} = \frac{2\pi}{\lambda} [\sin 2\theta_B \cos \phi, \sin 2\theta_B \sin \phi, \cos 2\theta_B]$, where θ_B is the Bragg angle and ϕ the azimuthal angle around the Debye-Scherrer ring. Thus, by using the Laue condition, ($\mathbf{G} = \mathbf{k} - \mathbf{k}_0$) we define \mathbf{G} , the diffracting reciprocal lattice vector, as $\mathbf{G} = \frac{2\pi}{\lambda} [\sin 2\theta_B \cos \phi, \sin 2\theta_B \sin \phi, \cos 2\theta_B - 1]$.

Consider a general vector \mathbf{A} connecting any two lattice points in real space. Under deformation of the crystal, the new vector connecting the two lattice points in the rotated coordinate system \mathbf{A}' will be related to \mathbf{A} by the deformation gradient-

$$\mathbf{A}' = \mathbf{F}\mathbf{R}\mathbf{A}, \quad (1)$$

where \mathbf{F} represents the deformation gradient and \mathbf{R} is the rotation matrix that takes our working coordinates to the sample coordinates. For simplicity, we will assume that only the diagonal components of the strain tensor are non-zero. Noting our target is tilted at 45° to the incoming

X-rays, we find-

$$\mathbf{F} = \begin{pmatrix} 1 + \varepsilon_{11} & 0 & 0 \\ 0 & 1 + \varepsilon_{11} & 0 \\ 0 & 0 & 1 + \varepsilon_{33} \end{pmatrix}, \quad \mathbf{R} = \begin{pmatrix} \frac{1}{\sqrt{2}} & 0 & -\frac{1}{\sqrt{2}} \\ 0 & 1 & 0 \\ \frac{1}{\sqrt{2}} & 0 & \frac{1}{\sqrt{2}} \end{pmatrix}, \quad (2)$$

where ε_{11} and ε_{33} are the components of the strain tensor that are transverse to and along the direction of shock propagation, respectively. However, reciprocal lattice vectors, and therefore \mathbf{G} , will transform with the transpose of the inverse of the real space transform \mathbf{F} [15]. Therefore, if we want to transform the \mathbf{G} vector corresponding to a region of compressed material back to its position before the grain was compressed, \mathbf{G}'_0 , we use $\mathbf{G}'_0 = \mathbf{F}^T \mathbf{R} \mathbf{G}$. By taking the modulus of both sides, $|\mathbf{G}'_0| = |\mathbf{F}^T \mathbf{R} \mathbf{G}|$, we obtain [15]-

$$\begin{aligned} \frac{\lambda^2}{d_0^2} &= \frac{1}{2} \left((1 + \varepsilon_{11})^2 + (1 + \varepsilon_{33})^2 \cos^2 \phi \right) \sin^2 2\theta_B \\ &+ \left((1 + \varepsilon_{33})^2 - (1 + \varepsilon_{11})^2 \right) \cos \phi \sin 2\theta_B (\cos 2\theta_B - 1) \\ &+ \frac{1}{2} \left((1 + \varepsilon_{11})^2 + (1 + \varepsilon_{33})^2 \right) (\cos 2\theta_B - 1)^2. \end{aligned} \quad (3)$$

4. Experimental Results

By replacing θ_B and ϕ with ψ using the equation $\cos \psi = \frac{1}{\sqrt{2}}(\cos \theta_B \cos \phi - \sin \theta_B)$, the formula above simplifies to-

$$(d/d_0)^2 = \varepsilon_{11}^2 + (\varepsilon_{33}^2 - \varepsilon_{11}^2) \cos^2 \psi. \quad (4)$$

By plotting $(d/d_0)^2$ against $\cos^2 \psi$, it is possible to extract the normal and transverse strain components, as shown in figure 3(a). These strains are then shown in figure 3(b). We see definite evidence for a strain anisotropy, suggesting that the plastic deformation has not completely relaxed the sample back to the hydrostat.

5. Results of Simulations

In an attempt to extract the stress components from the strains found above using the molecular dynamics (MD) package LAMMPS [16] and Sheng's embedded atom model (EAM) potential [17], a simulation of a 150x200x300 Å polycrystalline copper sample was performed. A model of the polycrystal was generated using the program fillatoms [18] and was then compressed by the strains found in the above section, and fed into the simulation to calculate the stress components, which were then used to find the shear stress, a measure of material strength. The results are shown in Figure 4, alongside gas gun data from Millett *et al.* [9] and explosive lens data from Bat'kov *et al.* [6]. A comparison of the datasets is given in the paper by Millett [9].

6. Summary

In summary, we have measured the strain anisotropy of a laser-shocked copper foil in the first long-pulse laser experiment using the MEC instrument at LCLS. By tilting the foil with respect to the incoming X-rays, the normals of different diffracting planes with a Debye-Scherrer ring will form different angles to the direction of shock propagation. This breaking of the symmetry of the experiment allows us to be sensitive to strain anisotropy by measuring the different shifts in the Bragg angle at different points on the Debye-Scherrer ring. By taking the extracted strains and putting them into an MD simulation, we inferred a shear stress (a measure of material strength) of up to 1.75 GPa at 32 GPa, which is reasonably consistent with previous gas gun work.

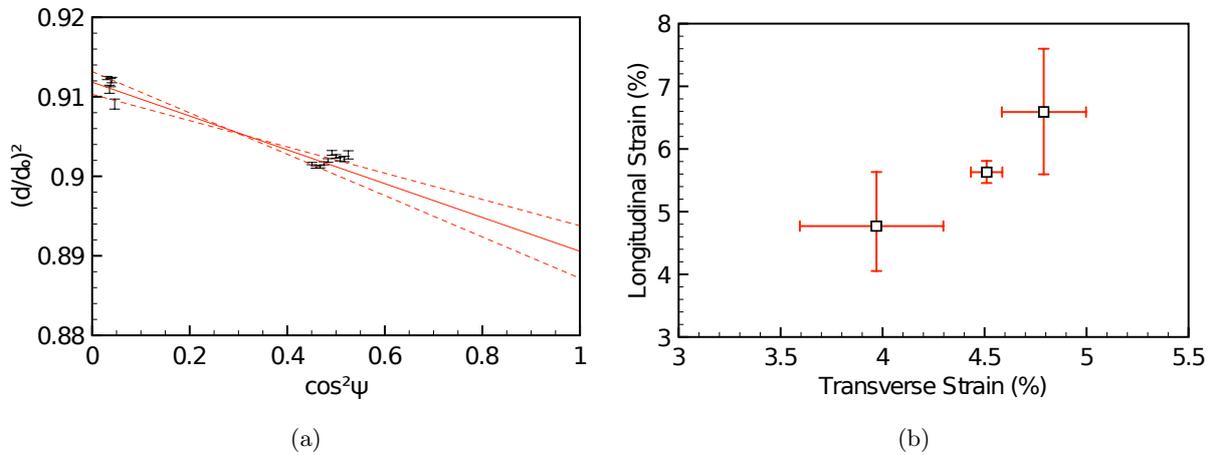


Figure 3. (a) A graph of $(d/d_0)^2$ against $\cos^2\psi$, where $d = \frac{\lambda}{2\sin\theta_B}$, d_0 is the original plane spacing and ψ is the angle between the diffracting grain normal and the direction of shock propagation. The central red line shows the best fit to the data, with the other two showing the error on the fit. (b) The longitudinal and transverse strains extracted by fitting the observed diffraction pattern.

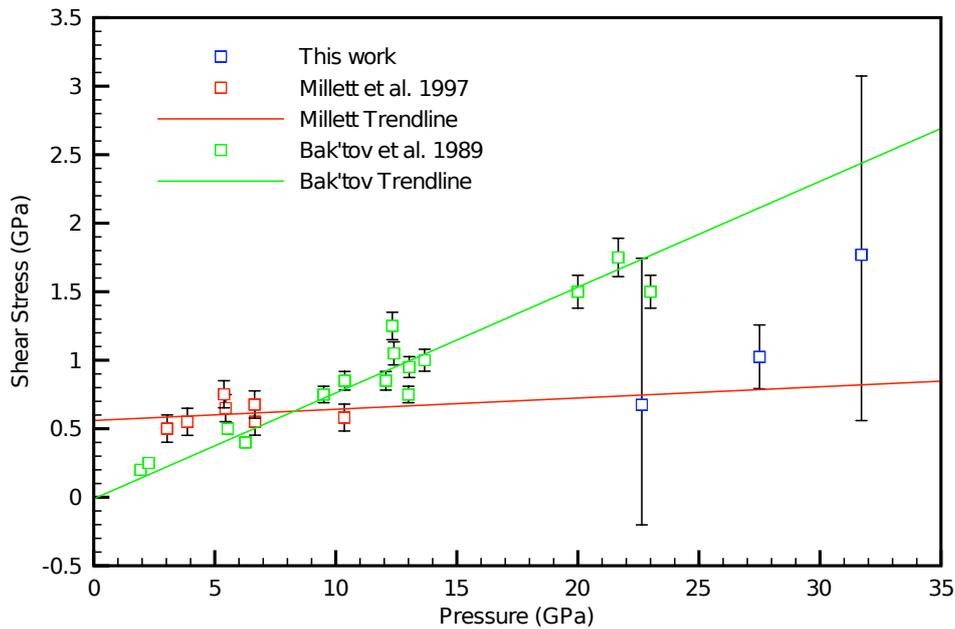


Figure 4. The inferred shear stress plotted against pressure gained from MD simulations. For comparison, gas gun data from Millett [9] and explosive lens data from Bat'kov [6] are also plotted.

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