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X-ray Diffraction From Shocked Crystals: Experiments and Predictions of Molecular Dynamics Simulations

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Abstract. When a crystal is subjected to shock compression beyond its Hugoniot Elastic Limit (HEL), the deformation it undergoes is composed of elastic and plastic strain components. *In situ* time-dependent X-ray diffraction, which allows direct measurement of lattice spacings, can be used to investigate such phenomena. This paper presents recent experimental results of X-ray diffraction from shocked fcc crystals. Comparison is made between experimental data and simulated X-ray diffraction using a post-processor to Molecular Dynamics (MD) simulations of shocked fcc crystals.

INTRODUCTION

Time-dependent *in situ* X-ray diffraction is an excellent tool for studying the structure of crystals under shock loading as it enables the investigation of the material of interest at the lattice level on picosecond time-scales. X-rays are scattered from the atoms in a crystal and interfere constructively when the well-known Bragg condition is satisfied:

$$\lambda = 2d_{hkl}\sin\theta_B, \qquad (1)$$

where λ is the x-ray wavelength, θ_B is the Bragg angle between the diffracted signal and the planes defined by indices (hkl), and d_{hkl} is the spacing of the crystal planes labeled by the indices (hkl) . Although shifts in the diffraction peak can be caused by dislocations, the angular shifts in the diffracted radiation are principally sensitive to the elastic strains within the crystal, and thus observation of compression of the lattice in directions both perpendicular and parallel to the shock front is expected when the crystal deforms plastically. Under conditions of uniaxial strain, where the elastic and plastic components of transverse strain are equal and opposite, the measurement of transverse elastic strain by X-ray diffraction gives a direct measure of plastic strain. The concept of the technique is illustrated schematically in fig. 1. An early, simple and well-known model for this lattice response was initially put forward by Smith [1], and later refined by Meyers [2], [3] and is illustrated in fig. 2.

Indeed, previous experiments, where laser-driven shocks have been driven into single crystals and diffraction from planes both parallel and perpendicular to the shock propagation direction observed, have demonstrated significantly different behaviour between laser-shocked silicon, and face-centred-cubic metals such as copper [4]. Diffraction from the shocked copper crystals during the compression shows that the crystal deforms in a manner apparently close to hydrostatic compression – rapid (sub-nanosecond) compression of the lattice in directions both parallel and perpendicular to the shock front is observed, consistent with the simple pictures of lattice deformation.

In addition, our theoretical understanding of the response of crystals to shock compression has been significantly advanced over recent years by the advent of molecular dynamics (MD) calculations which can simulate the response of tens of millions of atoms for tens of picoseconds. Such capabilities give rise to the possibility to simulate macroscopic (micron) sized crystals for time-scales similar to those used in the experiments briefly commented upon above. It is, therefore, of interest to compare the results of the diffraction experiments with the predictions of the MD calculations. To this end, a simple post-processor has been written for the MD simulations which produces diffraction patterns that can be compared with experimental data. We present below initial results from this post-processor that provide an indic-

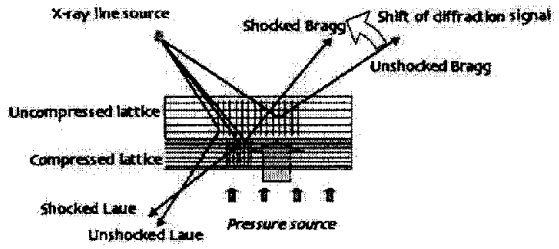

FIGURE 1. Bragg and Laue diffraction cases from perfect and compressed parts of a crystal. In the Bragg case, X-rays diffract from planes perpendicular to the shock direction. In the Laue case, diffraction is from planes parallel to the shock direction. In this way, both longitudinal and transverse strains in the crystal can be detected.

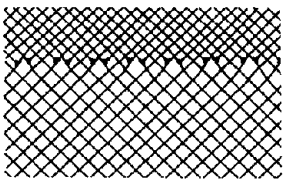

FIGURE 2. Interface formed by dislocations at the shock front, as modeled by Meyers [3]. This formation relieves shear stress present and a cubic lattice with reduced unit cell size is formed behind the shock front.

ation of the future work required to find good agreement between MD simulations and the experimental data referred to above.

EXPERIMENTS

The experiments which motivated the simulations described here have been described extensively elsewhere, [4] and thus we only provide a brief description here for completeness.

Experiments were conducted on the OMEGA laser to study the shock response of Cu to laser irradiation. A single crystal of Cu was shock loaded by nanosecond time-scale direct drive. The sample was probed by x-rays generated with high intensity beams incident on a thin metal foil. A nearly monochromatic source of x-rays was created. These x-rays were then diffracted from the lattice planes of the crystal and recorded on both static film and x-ray streak cameras. X-rays diffracted from the (002) planes of Cu parallel to the shock propagation direction in the crystal were recorded with one detector. X-rays diffracted from the orthogonal (020) planes of Cu were recorded with a separate detector. The foils were shock compressed to a peak pressure of approximately 200 kbar. X-rays diffracted from the (200) and (020) lattice planes showed a compression of apprimately 2.5-

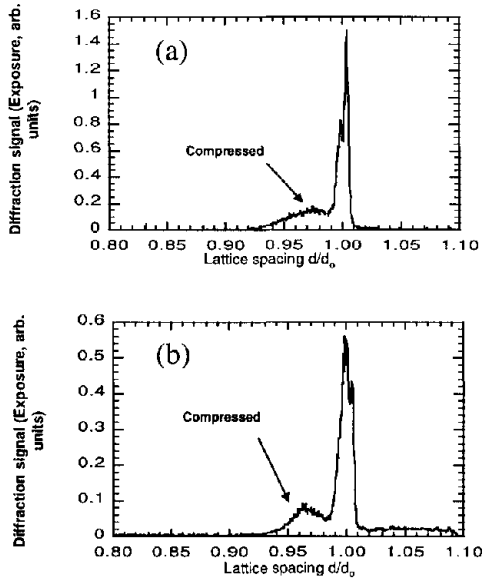

FIGURE 3. Experimental X-ray diffraction data for a thin laser-shocked Cu crystal for the conditions stated in reference [4]. Diffraction from planes with reciprocal lattice vector (a) parallel to shock propagation direction, and (b) perpendicular to shock propagation direction.

3%, as illustrated in fig.3. This data is consistent with close to 3-dimensional compression of the Cu. Since the foil is very thin (8 μm) compared to the transverse dimension (1 mm), this volumetric compression is a result of the local rearrangement of the lattice under compression due to the generation and propagation of dislocations. Given the time-scales of the experiment, we conclude that such rearrangement of the lattice can take place on sub-nanosecond time-scales which may, in the near future, be directly accessible to simulation by MD techniques.

COMPUTATIONAL ANALYSIS

Post-processing MD simulations to provide X-ray diffraction simulations that can be compared with the experimental data is conceptually very simple. The sample sizes used in the MD simulations are sufficiently small that, even in the absence of dislocations, the diffraction is kinematic (i.e. extinction can be ignored). Thus the intensity, I, of the diffracted radiation is given by

$$I = |\sum_j f_j \exp[\ \ i(\mathbf{r}_j \cdot \Delta\mathbf{k})]|^2, \qquad (2)$$

where f_j is the atomic form factor of the jth atom, $\mathbf{r}_j$ its position, and $\Delta\mathbf{k}$ the difference between the wavevectors of the incident and scattered radiation. This formula for the diffracted intensity also neglects the effects of absorption within the sample. Absorption can easily be taken into account by keeping track of the path length traversed by the X-rays scattered from a given atom, but again current simulated sample sizes make this unnecessary. In the simulations we provide below we make the assumption that the X-rays undergo specular reflection (by constraining $\Delta\mathbf{k}$ to always be parallel to a particular reciprocal lattice vector of interest of the unshocked crystal). This is an assumption we will comment upon later.

Various types of MD simulations modeling the behaviour of fcc crystals undergoing shock compression have been developed [5], [6]. This paper presents preliminary results of X-ray postprocessing of a simulation where the shock is generated using a piston, which consist of two layers of atoms moving at fixed velocity along the [001] direction. The piston specularly reflects any atoms reaching it. A shock wave then moves away from the piston at velocity $u_s \quad u_p$, where u_s is the planar shock velocity and u_p is the piston velocity. Periodic boundary conditions are applied in the lateral directions, with a free surface at the end of the sample far from the piston. The potential used is EAM [7].

Fig. 4 shows the simulated diffraction signal from a sample of size $50 \times 50 \times 200$ fcc unit cells. The diffracted intensity is plotted as a function of Δk, normalized to the length of the [001] reciprocal lattice vector. The pressure applied was 50GPa. The shock applied has no rise-time other than the intrinsic rise-time, and the snapshot taken is 7.81ps after the shock has started propagating, before it reached the rear surface. For clarity the contribution to the diffraction from unshocked part of the crystal has not been included. There are several important features to note from this figure. Firstly, as expected we observe diffraction peaks close to those positions corresponding to those miller indices allowed by the face-centred-cubic lattice (i.e. in this case, where we are simulating diffraction from planes with reciprocal lattice vectors that are parallel to the sides of the conventional unit cell, only even orders are observed). Secondly, and most importantly, it can be seen that a noticeable shift in the diffraction signal is mainly observed in the shock propagation direction - indeed in the second order no observable shift can be seen in the diffraction from planes with reciprocal lattice vectors perpendicular to the shock propagation, although some shift is seen in fourth order. If we infer elastic strains from the angular shifts of the diffracted radiation, then the second order diffraction implies a compression along [001] (the shock propagation direction) of 16.5±1.5%, and along [100] and [010] of 0±1.5%. Fourth order is more sensitive to compression in that the change in Δk is proportional to the order, however the signal is also noisier. For this order we infer 13±2% compression along [001], and 3.5±2% along [100] and [010].

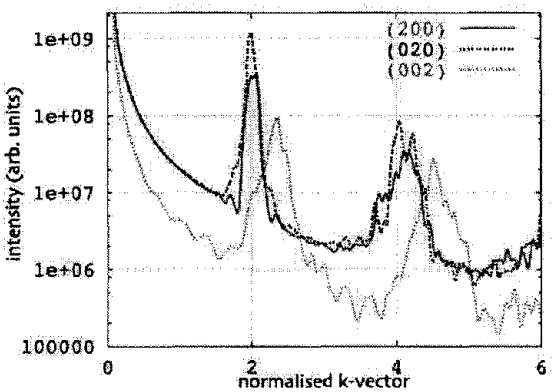

FIGURE 4. Diffraction Intensity (arbitrary units) as a function of Δk, normalized to the length of the [001] reciprocal lattice vector, for the conditions stated in the text. The diffraction signature of compression along the shock propagation direction is seen clearly, with significantly less compression in the two orthogonal directions.

Note that under the assumption of zero plastic dilatation the sum of the elastic strains in each of the three orthogonal directions should equal the volumetric compression. Thus, given the error bars associated with the simulated shifts in the diffraction features, the observed elastic strains inferred from the diffraction are consistent with the 18 % compression deduced directly from MD simulations. Note that although little shift in diffraction signals are seen corresponding to elastic strain orthogonal to the shock propagation direction, the dislocation density in the shocked region of the crystal is high.

We note that several forms of diffuse scattering are, or may be, occurring, and these will need to be taken into account in future simulations by relaxing the constraint that $\Delta\mathbf{k}$ be parallel to the relevant reciprocal lattice vector. The first form of non-specular diffraction that has not yet been taken into account is simply due to the finite divergence of the diffracted radiation. In conventional dynamical diffraction theory from perfect crystals one assumes non-divergent beams as the crystals of interest are large: the divergence is inversely proportional to the number of unit cells along the surface of the crystal, and the rocking curve width inversely proportional to the effective number of scatterers parallel to the relevant reciprocal lattice vector. However, owing to the finite number of atoms within this simulation, the outgoing beam will

diverge with a divergence inversely proportional to the number of atoms perpendicular to the reciprocal lattice vector. Indeed, this is why the scattered intensity from the planes associated with a reciprocal lattice vector parallel to the shock propagation direction differs from that in the two orthogonal directions. These effects will become less important as the scale of the simulations increases. More importantly, further reduction in the integrated intensity will occur due to the local rotations of the reciprocal lattice vectors which are associated with finite rotations of small regions of the lattice during the generation of dislocations. We also expect some degree of diffuse scattering far due to thermal effects (the Debye-Waller effect), although for the temperatures present here we do not expect this to be a large effect. These effects will be taken into account in future simulations, and clearly it will be important to investigate the effect of diffuse scattering and sample size on the error bars of the elastic strains deduced from the shifts in the simulated diffraction signals.

CONCLUSION AND FUTURE WORK

Results of the X-ray diffraction simulating post-processor applied to the MD simulations presented in this paper indicate that compression of an fcc metal lattice mainly occurs along the shock propagation direction on time-scales of a few picoseconds, even though considerable dislocations densities are produced by homogeneous nucleation at the shock front. Experimental data shows significant compression in all directions on time-scales of a few hundred picoseconds [4]. Thus it would be of interest in the future to apply a similar analysis to larger MD simulations, run over considerably longer time-scales.

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