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Femtosecond measurement of shock wave driven twinning and lattice dynamics

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Understanding the deformation and associated defects created by shock waves is crucial in a wide range of fields, such as planetary formation and asteroid impact sites¹⁻³, formation of interstellar dust clouds⁴, ballistic penetrators⁵, spacecraft shielding⁶, and ductility in high-performance ceramics⁷. Shock waves in solid materials can cause extreme damage and deformation. Twinning and dislocation-slip are the basic mechanisms of plastic deformation at the lattice level, yet diagnosing the active mechanism *in-situ* has been elusive. Methods for characterizing lattice defects have typically been limited to ex-post-facto experiments⁸⁻¹¹, which examine the microstructures of samples and are complicated by post-shock annealing¹² and reverberations. In addition measurements have been limited to relatively modest pressures. *In-situ* X-ray diffraction (XRD) experiments have provided new insights into dynamic material behavior¹³ but are only starting to be applied to plasticity during shock compression¹⁴⁻¹⁷, and have yet to provide detailed insight into competing deformation mechanisms. Here we present XRD experiments with femtosecond resolution that apply crystallographic techniques to capture *in-situ*, lattice-level pictures of the microstructural processes driving shock deformation. Tantalum, an important material for high-energy density physics applications, serves as a model body-centered cubic (BCC) material. For shock-compressed Ta, where simulations¹⁸⁻²⁰ and previous experiments⁸⁻¹² provide conflicting information on the dominant mechanism, we report twinning and related lattice rotation occurring on the time scale of tens of picoseconds. Despite a common association between twinning and strong shocks²¹, we also find a transition from twinning to slip-dominated plasticity at high pressure (<150 GPa), a regime recovery experiments cannot accurately access. As the role of *in-situ* diffraction for studying shock waves and other high-rate phenomena continues to grow, the texture analysis techniques demonstrated here will be useful in studying a broad range of processes induced by plasticity.

40 Prior *in-situ* experimental measurements of lattice evolution during shock measurements
41 have not had sufficient time or spatial resolution to make critical direct comparisons to theory.
42 The need for a more direct comparison to theory is evident since, even for a well-studied BCC
43 metal such as Ta, existing experiments can disagree on the operative deformation mechanism,
44 twinning versus dislocation generation and transport (slip). Ta samples show significant twinning
45 at shock pressures above ~ 50 GPa⁸⁻¹¹, whereas previous *in-situ* diffraction experiments have not
46 registered a signal indicative of twinning at these pressures^{16,17}. While the wide difference in
47 timescales makes comparing between experiments difficult, large-scale molecular dynamics
48 (MD) simulations in strongly shocked Ta also disagree. The deformation mechanism varies from
49 slip¹⁸ to twinning¹⁹ to a combination of the two mechanisms²⁰, depending on which interatomic
50 potential is used in the simulation. In this study the new capabilities offered by x-ray free-
51 electron laser sources are used to make direct comparisons to molecular dynamics simulations.

52 Here we use temporally resolved, *in-situ* x-ray diffraction to probe shocked polycrystalline
53 Ta, providing direct characterization of the mechanism and time scale of the ultrafast
54 deformation for shock pressures from 10 GPa to shock melt (~ 300 GPa). The experiments
55 were performed at the Matter in Extreme Conditions end station of the Linac Coherent
56 Light Source, and a sketch of the setup is shown in Figure 1. Probed during the initial
57 shock transit through the Ta, the diffraction patterns have signal from both driven and
58 undriven material. The material transitions rapidly to the shocked state. The low signal
59 level between driven and undriven lines implies a fast transition, occurring on timescales of
60 < 100 ps for the highest pressures ($> 10^9$ s⁻¹ strain rate).

61 The Ta foil was fabricated by vapor deposition, which creates a fiber-like texture where
62 the grains share a common crystallographic direction along the sample normal ([110] in
63 this case) but are oriented randomly in the transverse direction. Having a known, highly-
64 textured starting material means new orientations created by deformation can be readily
65 observed in the diffraction data²². Twinning and slip produce distinct changes to the
66 texture of the Ta sample that can be used to determine the active mechanism for
67 deformation during shock compression. Both twinning and slip can cause a rotation of the
68 lattice when the sample is laterally confined during shock compression, since a lattice
69 rotation is needed to maintain the geometry of uniaxial compression²³. However, twinning
70 also produces an additional mirror rotation in the twin grain.

71 Due to the highly-textured structure of the Ta foil, only certain spots are visible on
72 the powder Debye diffraction ring. The azimuthal positions of the spots correspond to the
73 angles of the respective diffraction planes. Figure 2 shows an example diffraction pattern
74 taken prior to shock loading. Contours of constant χ -angle, the angle between the
75 sample surface and the diffraction plane, are overlaid on the figure (constant 2θ is shown
76 with dashed lines). Two sketches of a Ta unit cell demonstrate that the $\{110\}$ planes will
77 occupy angles of 60° or 90° relative to the sample plane. The $\{110\}$ diffraction ring has
78 texture spots at corresponding χ of 60° and 90° (marked with green arrows). Similar
79 reasoning accounts for the azimuthal positions of texture spots for the other diffraction
80 rings.

81 Texture changes are evident in the driven data for a wide range of shock pressures, as
82 shown in the example diffraction patterns in Figure 3. Comparing χ for the compressed
83 and uncompressed spots reveals the texture change during shock compression. In Fig. 3a
84 (23 GPa), the d-spacing, the distance between lattice planes, decreases in the driven

85 portion of the sample and the diffraction lines move to higher 2θ angles, but no change in
86 χ is observed. At 46 GPa (Fig. 3b), a new set of spots forms on the compressed Debye
87 ring, marked with red arrows, that correspond to an orientation for twinning across a
88 $\{112\}$ mirror plane (the dominant twin plane in BCC Ta). In the higher-pressure data (Fig
89 3c and d), the $\chi = 60^\circ$ spot is elongated azimuthally; the texture spot spans the detector
90 gap in Fig. 3c and even splits into two spots (Fig. 3d). These changes in χ correspond to
91 lattice rotation, where the rotation moves some planes to higher χ and other planes to
92 lower χ . In contrast, no shift in χ is expected for the $\chi = 90^\circ$ spot since the rotation occurs
93 in this plane.

94 The amount of texture change is plotted in Fig. 4a (twinning) and 4b (lattice rotation).
95 The twin fraction is minimal (consistent with zero) below 25 GPa, and increases with
96 shock pressure in the 25-75 GPa regime, reaching a twin fraction of $\sim 30\text{-}40\%$ at 75 GPa.
97 Surprisingly, even though twinning is often associated with strongly driven metals, at
98 higher shock pressures ($>150\text{-}175$ GPa), the twin fraction falls to low levels ($< 10\%$). The
99 lattice rotation in Fig. 4b can be due to either slip or twinning and is found to increase
100 monotonically. Thus the decrease in twin fraction observed above ~ 150 GPa indicates a
101 transition to slip-dominated plasticity.

102 Figure 4c plots the equivalent plastic strain associated with twinning, γ_{Tw}^P , orange
103 triangles) and lattice rotation, γ_{Ro}^P (purple diamonds), while the plastic strain from slip can
104 be determined from the difference between γ_{Ro}^P and γ_{Tw}^P . The total strain for a given
105 shock is shown by γ_{Hu}^P (black line), the equivalent plastic strain on the Hugoniot. If either
106 mechanism dominates the response, the plastic strain from that mechanism will be a large
107 fraction of γ_{Hu}^P . While the onset of twinning occurs at 25 GPa, the plastic response is twin-
108 dominated in the 40-80 GPa range. Above 150 GPa, the plastic strain from twinning falls
109 well below the Hugoniot, indicating the plastic response becomes slip-dominated.

110 The trend for the lattice rotation follows the Hugoniot with a small offset. There is a
111 difference of 4% strain between the two curves, $\gamma_{Hu}^P - \gamma_{Ro}^P \sim 0.04$, indicated by the dashed
112 line in Fig 4c. In Fig. 4 b-d, the $\chi = 90^\circ$ spots of the driven (110) Debye rings show
113 some azimuthal broadening. Since, as noted above, slip along $\{112\}$ planes does not
114 affect the (110) $\chi = 90^\circ$ planes, this broadening is an indication of a small amount of slip
115 along other planes and likely accounts for the 4% difference.

116 Phase transformations can also create texture changes and should be considered as a
117 possible response to shear strain²². While density functional theory indicates the BCC phase
118 is the stable phase for pressures up to 700 GPa and temperatures up to the melt line²⁴, the
119 plastic deformation of shock compression may drive a transition to the omega phase, as
120 indicated by recovery and MD simulations^{2,25}. Our data does not show significant amounts
121 ($>0.1\%$) of omega phase (see ED Fig. 1 & ED Fig. 2) or any other solid phase apart from
122 BCC for all pressures, from ambient up to melt at ~ 300 GPa.

123 We performed MD simulations using the Ravelo potential²⁶, simulating a Ta single crystal at
124 300 K shocked along the [110] direction. Figure 4a shows the simulated twin fraction and
125 Fig. 4b shows the simulated lattice rotation as a function of pressure (black circles). The
126 MD simulations show the onset of twinning occurring at similar pressures as the data,
127 although the decrease in twin fraction occurs at much lower pressure in the simulations.
128 The simulated lattice rotation is in good agreement with the data in the 75-200 GPa

129 pressure range.

130 The quantitative measurements provided by XRD allow direct comparison to simulations
131 and modeling. Complex constitutive models like the Livermore Multiscale Model²⁷ begin
132 with the assumption that plasticity is dislocation based. Even in the simplified case studied
133 here, where no phase transformations occur and loading is restricted to a single crystal axis,
134 the dominant deformation mechanism transitions multiple times, from minimal twinning to
135 twinning-dominated to slip-dominated, as the shock pressure is increased. Approaching
136 similar timescales to simulations and exploring high pressure regimes that are difficult to
137 observe and interpret in *ex-post-facto* experiments, these results highlight the importance
138 and growing role of *in-situ* experiments in studying materials in extreme conditions.

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142

143 **Fig. 1. Experimental configuration used at the MEC end station at LCLS for shock**
144 **compression and *in-situ* x-ray diffraction.** A shock wave is driven by laser ablation
145 into a plastic ablator and Ta foil with a (110) fiber texture. The shock compressed Ta is
146 probed by the LCLS beam and the resulting diffraction patterns are collected on an
147 array of CSPADs.

148

149 **Fig. 2. Example x-ray diffraction data from (110) fiber textured Ta prior to shock**
150 **loading.** A sketch of the diffraction geometry describes 2θ , the Bragg angle, and χ , the
151 angle between the sample normal and diffraction plane normal. For a sample with (110)
152 texture, {110} planes have $\chi=60^\circ, 90^\circ$. The diffraction data have corresponding {110}
153 texture spots at those χ angles, marked with green arrows. Solid curves indicate
154 constant χ corresponding to specific lattice planes [for {110}: $\chi=60^\circ, 90^\circ$; for {200}:
155 $\chi=90^\circ, 45^\circ$; for {211}: $\chi=73^\circ, 54.7^\circ$]. The dashed curves indicate constant 2θ .

156

157 **Fig. 3. Diffraction patterns from Ta shock compressed at (a) 19, (b) 44, (c) 108,**
158 **and (d) 169 GPa.** The driven spots appear at higher diffraction angle, which appears
159 as a shift upwards in this view. Lines of constant χ -angle are shown at 60° and 90° to
160 illustrate the reorientation of the texture spots in the driven Ta. At 23 GPa (a) no
161 reorientation is observed, since the driven and undriven diffraction spots have the
162 same azimuthal location. At 46 GPa, new texture spots arise corresponding to
163 twinning across a {112} mirror plane plus an additional 2.5° rotation. At higher
164 pressures, (c) and (d), large lattice rotations are observed, causing the $\chi=60^\circ$ (110)
165 spot (marked with red arrows) to elongate and then split.

166

167 **Fig. 4. Twin fraction, rotation and plastic strain computed from the diffraction data.**
168 (a) Twin fraction as a function of shock pressure measured by the ratio of texture spots
169 (orange triangles) and from MD simulations (black circles). (b) Lattice rotation angle from
170 XRD (purple diamonds) and from MD simulations (black circles). (c) Equivalent plastic
171 strain inferred from lattice rotation (purple diamonds) and twin fraction (orange triangles).
172 The equivalent plastic strain on the Hugoniot is plotted (black line). The rotation data
173 follows the Hugoniot line minus 4% strain (dotted line). Twinning accounts for the
174 majority of the Hugoniot plastic strain in the 40-80 GPa range (pink shaded region is a
175 guide to the eye), but falls well below the Hugoniot above 150 GPa (blue shaded region),
176 indicating the response is slip dominated.

177

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290 **Methods**

291 The experiments were performed at the Matter in Extreme Conditions (MEC) end station of
 292 the Linac Coherent Light Source (LCLS). The Ta structure was probed while under shock
 293 compression and the resulting diffraction patterns were recorded on several Cornell-SLAC
 294 hybrid Pixel Array Detectors (CSPADs)²⁷, as shown in Fig. 1. A planar shock was driven by
 295 direct laser ablation from frequency doubled (527 nm) laser beams, using a 5-10 ns square pulse.
 296 Hybrid phase plates were used to modify the focal spot. These plates use surface features to shift
 297 the phase of the light, creating focal spots of 100 μm, 150 μm or 250 μm diameter with “top-hat”
 298 intensity profiles. Each target consisted of a 50 μm thick kapton ablator glued to a 6 μm thick
 299 foil of Ta. The ablator thickness (several times larger than the sample) prevents reverberations
 300 from entering the Ta before the initial shock has passed through the sample. The thickness of the
 301 ablator also helps to smooth out spatial variations in drive created by speckle pattern of the laser
 302 spot. The Ta foil was fabricated by vapor deposition onto a SiO₂ substrate at a temperature
 303 (~450°C) sufficient both to create the body-centered cubic (BCC) α-Ta phase and to delaminate
 304 the Ta from the substrate upon cooling. Small amounts (<0.5%) of β phase are also present in
 305 some of the Ta samples.

306 A Velocity Interferometer System for Any Reflector (VISAR) diagnostic was used to
 307 monitor the free surface velocity of the Ta during the shock compression and release and
 308 help set the diffraction timing. The optical streak camera system (line VISAR) was used to
 309 confirm the planarity of the drive. The VISAR images show the planar driven region
 310 approximately corresponds to the size of the phase plates, and this planar region larger than the
 311 20-μm x-ray beam. An example of the VISAR data for a 130 GPa shock is shown in ED Fig 3.

312 The LCLS x-ray beam was tuned for 9.6 keV photons in a 50 fs pulse. The diffraction angles
 313 were calibrated using a powder sample of LaB₆ and CeO₂ standards. The calibrated detector
 314 positions were then used to measure the diffraction angles from the Ta samples. The sample
 315 orientation is determined by fitting the χ of the spots of the ambient diffraction pattern. The
 316 amount of lattice rotation can then be determined from the χ of the driven spots. A single
 317 rotation from the original [110] orientation is determined that best fits the measured χ of the
 318 driven spots. The diffracted intensity was adjusted to account for polarization of the x-ray beam
 319 using the following factor:

320
$$P = (\cos^2 \phi * \cos 2\theta)^2 + \sin^2 \phi \quad P = [\cos^2 \phi * \cos(2\theta)]^2 + \sin^2 \phi$$

321 where φ is the azimuthal angle for diffracted x-rays. The pressure produced by the shock loading
 322 was calculated from the d-spacing measured by diffraction assuming BCC structure.
 323 Specifically, the shock pressure was found using the shock speed data from Mitchell and
 324 Nellis²⁸, $U_s = C_0 + S * U_p$ where $C_0 = 3293$ m/s and $S=1.307$, which together with the Rankine-
 325 Hugoniot equations, give the following expression for the shock pressure²⁹:

326
$$P_x = \rho_0 C_0^2 \frac{1 - (d/d_0)^3}{1 - S[1 - (d/d_0)^3]} \quad P_x = \rho_0 C_0^2 (1 - [d/d_0]^3) / (1 - S[1 - (d/d_0)^3])$$

327 where ρ₀ is the initial density and d_0 and d are the initial and compressed lattice spacings. Since
 328 the error in determining the peak positions in the diffraction data is small, the main sources of
 329 uncertainty in determining the pressure are the spatial and temporal non-uniformity in the drive.
 330 These non-uniformities will not significantly change the average pressure, but may affect the
 331 range of pressures present in the sample. The VISAR records show that the drive is steady prior

332 to the first reverberation. Some spatial non-uniformity is present but is small over the 20 μm
 333 region probed by the x-ray beam. Based on uncertainty in fitting the peak positions, the pressure
 334 uncertainty is +/- 3 GPa.

335 For high pressure shots, where the driven diffraction pattern had the diffuse signal indicative
 336 of a liquid sample, the pressure could not be reliably measured from the diffraction data. The
 337 pressure estimate from the laser ablation intensity is consistent with previous measurements of
 338 the shock melting pressure, $\sim 300 \text{ GPa}^{30}$.

339

340 **Expected signal if omega phase (ω) were present**

341 We have analyzed the diffraction data for the presence of omega (ω) phase. This phase has
 342 been observed in shock-recovered Ta samples by Hsiung and Lassila² and Lu et al.²⁵. Hsiung
 343 provides the following relationship between the BCC and hexagonal lattice parameters:

$$344 \quad a_{\omega} = \sqrt{2}a_{\alpha}, c_{\omega} = \frac{\sqrt{3}}{2}a_{\alpha} \quad a_{\omega} = \sqrt{2}a_{\alpha}, c_{\omega} = \sqrt{3}/2a_{\alpha}$$

345 where a_{α} is the lattice parameter of the BCC phase, and c_{ω} and a_{ω} correspond to the omega
 346 phase. Lu et al. were able to match their TEM diffraction pattern using these lattice parameters.
 347 Lu et al. reported observing ω phase in samples that are recovered following shock loading to
 348 $\sim 70 \text{ GPa}$. In ED Fig 1. we show our driven data for a 73 GPa shock and indicate the expected
 349 peak positions for the hexagonal structure as well as the “rumpled” hexagonal structure reported
 350 by Hsiung and Lassila.

351 Since the initial ta structure is highly textured, the transformed omega phase may also be
 352 highly textured. Lu et al. state that they observed the same orientation relationship between the
 353 matrix and omega phase as Hsiung and Lassila, namely that the $(111)_{\text{m}} // (0001)_{\omega}$ and $[110]_{\text{m}} //$
 354 $[1000]_{\omega}$ where m refers to the matrix BCC phase and ω is the transformed omega phase.

355 Using this relationship, we show in ED Fig. 2. the azimuthal positions of the texture spots for the
 356 omega phase (white lines). For simplicity, the rumpled structure is not shown. The detectors
 357 cover multiple spots where the omega peaks would be expected given the previously reported
 358 orientation relationships yet no signal was observed.

359

360 **Twin fraction calculation**

361 We have measured the twin fraction by comparing the intensity of texture spots
 362 corresponding to the twin orientation with the intensity of other driven and undriven texture
 363 spots. In addition, the differing multiplicities of various planes associated with specific χ must
 364 be considered. For example, a unit cell in the twin orientation will have twice as many planes for
 365 $\chi = 90^{\circ}$ than $\chi = 70.5^{\circ}$, so that the twin fraction is given by,

$$366 \quad f_{\text{Tw}} = \frac{2 * I_{\text{Drvn}(110)\chi=70.5}}{I_{\text{Drvn}(110)\chi=90}} \quad f_{\text{Tw}} = (2 * I_{\text{Drvn}(110)\chi=70.5}) / (I_{\text{Drvn}(110)\chi=90})$$

367 where I refers to the integrated intensity of the specified diffraction spot. This equation assumes
 368 azimuthal symmetry, since the two spots being compared have different azimuthal positions.
 369 The azimuthal variation of the (110) fiber texture was not known prior to the experiments, so the
 370 variation is a source of uncertainty in our measurements. Pole figure measurements were
 371 performed on remaining samples, and azimuthal lineouts, normalized relative to the azimuthal
 372 average, follow a roughly sinusoidal pattern with 180° rotational symmetry and variation in
 373 intensity of +/- 30%.

374 Similar calculations were performed for the (200) $\chi=76.3^\circ$ and (211) $\chi=61.3^\circ$ twin spots, and
 375 the results were averaged to arrive at the final twin fraction values. The error bars were set to the
 376 standard deviation of the measurements using the three lines. For shock pressures in the 50-150
 377 GPa range, some of the driven twin spots fall into gaps between the detectors, limiting
 378 measurements in this pressure regime. The error was assumed to be 50% when twin spots were
 379 measured on only one diffraction line, which reflects the uncertainty in the azimuthal texture in
 380 both the twin spot and the reference spot.

381 We can quantitatively determine the role of twinning in the overall plastic response by
 382 comparing the plastic strain from twinning, γ_{Tw}^P , to the total plastic strain imparted by the
 383 shock, γ_{Hu}^P . Eqs. (1) and (2) give the two plastic strain quantities:

$$384 \quad \gamma_{Tw}^p = f_{Tw} * \epsilon_0 * M \quad \gamma_{Tw}^P = f_{Tw} * \epsilon_0 * M \quad (1)$$

$$385 \quad \gamma_{Hu}^p = 2/3 \ln\left(\frac{V_0}{V}\right) \quad \gamma_{Hu}^P = 2/3 \ln(V_0/V) \quad (2)$$

386 where f_{Tw} is the twin fraction, ϵ_0 is the eigenstrain, and M is the Schmid factor (for
 387 [110] loading, $\epsilon_0=0.707$ and $M=0.471$), and V_0 and V are the specific volumes in the
 388 ambient and shocked conditions respectively^{31,21}. We have taken the residual elastic shear
 389 strain to be negligible due to the absence of diffraction ring distortion (discussed below),
 390 implying full plastic relaxation of the shear stress on the ultrashort time scale of the
 391 experiment.

392 The {112} planes serves as the dominant plane for both slip and twinning, meaning both
 393 mechanisms will produce similar lattice rotations. The observed rotation, found by fitting
 394 the azimuthal position of the diffraction spots, provides a measurement of the plastic strain
 395 caused by both mechanisms, γ_{Ro}^p , as shown by Eq (3),

$$396 \quad \gamma_{Ro}^p = \ln\left(\frac{\cos \chi}{\cos \chi_0}\right) \quad \gamma_{Ro}^P = \ln(\cos \chi / \cos \chi_0) \quad (3)$$

397 where χ_0 and χ correspond to the initial and final angles between the slip plane normal
 398 and the compression direction.

399

400 How the plastic strain is calculated as a function of rotation

401

402 Here we discuss the relation between the plastic strain and the rotation that is measured.
 403 Throughout this analysis we assume that the residual shear stress in the shocked Ta is zero or so
 404 small that it can be neglected. This assumption, justified by the ring distortion analysis in the
 405 next section, allows us to impose that the elastic strain in the shocked state is strictly hydrostatic.

$$406 \quad \epsilon_x^e + \epsilon_y^e + \epsilon_z^e = \ln\left(\frac{V}{V_0}\right) \quad \epsilon_x^e + \epsilon_y^e + \epsilon_z^e = \ln(V/V_0)$$

$$407 \quad \epsilon_x^e = \epsilon_y^e = \epsilon_z^e = 1/3 \ln\left(\frac{V}{V_0}\right) \quad \epsilon_x^e = \epsilon_y^e = \epsilon_z^e = 1/3 \ln(V/V_0)$$

408 where ϵ_x^e is the elastic strain in the shock direction and ϵ_y^e and ϵ_z^e are the transverse elastic
 409 strains. Similarly, we assume the plastic strains do not change the volume

410 $\epsilon_x^p + \epsilon_y^p + \epsilon_z^p = 0$ $\epsilon_x^p + \epsilon_y^p + \epsilon_z^p = 0$

411 That uniaxial compression of crystals produces a rotation of the lattice is well known, and the
412 rotation as function of compression is²³

413 $\epsilon_x^p = \ln\left(\frac{\cos \chi_0}{\cos \chi}\right)$ $\epsilon_x^p = \ln(\cos \chi / \cos \chi_0)$

414 where χ and χ_0 give the angle between the slip plane normal and the compression direction and
415 ϵ_x^p is the plastic strain along the compression direction.

416 Since the material is laterally constrained and compressed along the high symmetry [110]
417 direction, we assume that, while individual grains may expand or contract in different transverse
418 directions, the area of each grain transverse to the sample normal will remain the same. Thus we
419 find that the total strain in the two transverse directions remains the same:

420 $\epsilon_y^e + \epsilon_y^p + \epsilon_z^e + \epsilon_z^p = 0$ $\epsilon_y^e + \epsilon_y^p + \epsilon_z^e + \epsilon_z^p = 0$

421

422 $\epsilon_x^p = \epsilon_y^e + \epsilon_y^p = \frac{2}{3} \ln\left(\frac{V}{V_0}\right)$ $\epsilon_x^p = \epsilon_y^e + \epsilon_y^p = 2/3 \ln(V/V_0)$

423 therefore

424 $\frac{\cos \chi_0}{\cos \chi} = \left(\frac{V}{V_0}\right)^{2/3}$ $\cos \chi_0 / \cos \chi = (V/V_0)^{2/3}$

425 $\gamma_p = -\frac{2}{3} \ln\left(\frac{V}{V_0}\right) = \ln \frac{\cos \chi}{\cos \chi_0}$ $\gamma_p = -2/3 \ln(V/V_0) = \ln(\cos \chi / \cos \chi_0)$

426

427 For the (211) slip plane, $\chi_0 = 54.7$

428

429 Residual Elastic Shear Stress

430 We have analyzed the Debye rings for deviations from circularity that would indicate shear
431 stress in the shocked state. When using the Hugoniot jump conditions to calculate the pressure
432 from the measured d-spacing, or when calculating the equivalent plastic strain, we have assumed
433 that the residual elastic shear stress is near zero. Residual elastic shear strain would mean that the
434 strain is anisotropic, with higher elastic strain in the compression direction (shock propagation
435 direction) and less in the transverse direction. This anisotropic elastic strain would manifest as a
436 Debye-ring distorted by varying d-spacing, making it appear elliptical³². With no shear strain, the
437 Debye-ring has constant 2θ . As shear strain increases, the driven rings move to higher 2θ for
438 low χ (right side of the diffraction pattern) and to lower 2θ for high χ (left side of the diffraction
439 pattern). While 0% strain produces a good fit to the data, small shear strains may also match.

440 We find at 2-3% shear strain, the data no longer provides a good match. Thus the assumption
441 that the elastic shear strain is negligible is likely valid.

442

443 **Molecular Dynamics Simulations**

444 Molecular dynamics simulations were performed using the LAMMPS code³³ and the Ravelo
445 potential²⁶ for 100x100x800 unit cells (330x330x2644 Å). A portion of the end of sample is used
446 as a piston. That section is “frozen” so that it won't respond to external forces and then ramped
447 up to a set velocity to drive the shock. By performing a 3D Fourier Transform³⁴ of the
448 plastically deformed region behind the shock front, we can find the average rotation of the
449 crystal lattice about the [1-10] axis. The Per Atom Structure Factor (PASf) code¹² was used to
450 determine the twin fraction in the same region.

451

452 **Data Availability.** The datasets generated and analyzed during the current study are available
453 from the authors on reasonable request.

454

455

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485 ED Fig. 1. Diffraction angle (2θ) plotted vs. x-ray counts averaged over 2π in azimuthal
486 angle for a shock pressure of 73 GPa. The indices of the ambient pattern, matching BCC
487 with lattice parameter of $a=3.308 \text{ \AA}$, are marked in green. The driven pattern (black dots)
488 match BCC with $a=3.051 \text{ \AA}$. Indexed peak positions for the hexagonal phase are marked with
489 red lines, and peaks corresponding to the “ruffled” structure observed by Hsiung & Lasilla²
490 are marked with an asterisk. Both BCC and omega phase overlap the observed peaks, but
491 additional peaks are expected for the omega phase that are not observed.

492 ED Fig. 2. Diffraction pattern for 73 GPa shock showing raw data (left) and overplotted with
493 BCC and omega phase (right). The ambient BCC phase is marked with blue. Compressed and
494 rotated BCC phase is marked in black, and the expected positions of the omega phase are marked
495 in white.

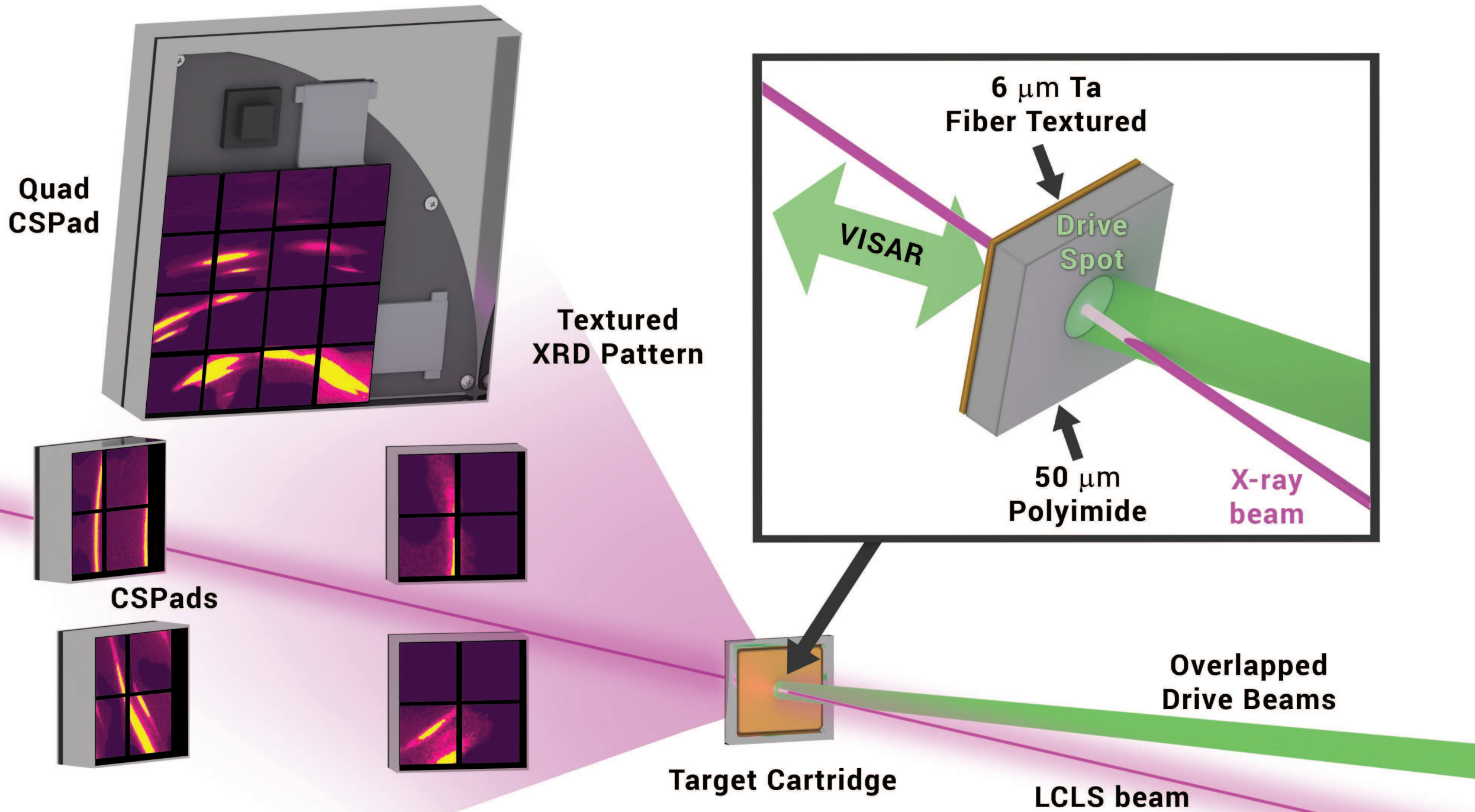
496

497 ED Fig. 3. Example VISAR streak image for a 130 GPa shot using a $250 \mu\text{m}$ phase plate. The
498 planar region of the shock, roughly similar in dimensions to the phase plate size, breaks out of
499 the Ta free surface at 7.4 ns. The x-ray beam probes a $20 \mu\text{m}$ region in the center of the planar
500 shock.

501

502

503



Sample Normal

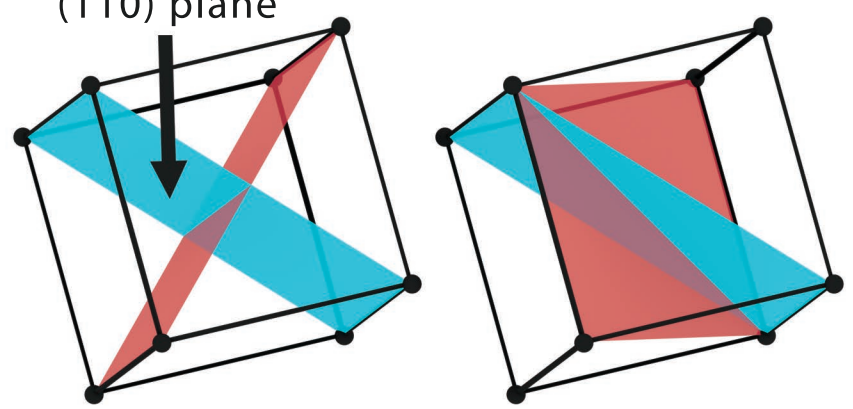
Plane Normal

C

2q

(110) Texture

Sample || to (110) plane



C=90°

C=60°

