Polychromatic microdiffraction analysis of self-organization in shock deformed single crystals

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Abstract
A 3D spatially resolved X-ray diffraction method - with a sub micrometer-resolution together with SEM and OIM analysis are applied to understand the arrangements of voids, geometrically necessary dislocations and strain gradient distributions in samples of Al (123) and (001) Cu single crystals shocked to incipient spallation fracture. We describe how geometrically necessary dislocations and the effective strain gradient alter white beam Laue patterns of the shocked materials. We show how to quantitatively determine the type and density of geometrically necessary dislocations in shock - recovered Al and Cu samples initially oriented for single and for multiple slip.

Keywords: dynamic fracture, voids and inclusions, nondestructive evaluation, characteristics, stress relaxation, shock waves

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1.**Introduction**

Wave propagation, in particular shock wave propagation, has proven to be a powerful tool for probing the mechanical behavior of materials under dynamic loading.[1] With increasing shock pressure, a change in material response occurs from elastic deformation to plastic deformation.[2 - 4] Strong shock waves produce an enormous number of dislocations at the shock front. However, the mechanisms of dislocation production, the density of dislocations, and the processes of dislocation annealing at later times to form dislocation structures are largely unknown.

The most reliable information to date on shock deformation has come from recovery experiments. In these experiments, a sample is shocked as with an impact from a light gas gun and then captured to minimize further deformation. Both peak shock pressure and pulse duration increase the residual dislocation density and resultant field strength. TEM studies reveal dense dislocation networks with cell sizes deceasing with increasing shock pressure.[5] Here, we compliment these studies with white (polychromatic) microbeam x-ray diffraction. The diffuse scattering surrounding the diffraction peaks results from the mesoscale dislocation arrangements inside the material. Single crystal Al (123) and Cu (100) were shocked to incipient spallation fracture and carefully recovered. The spall plane is characterized by a region of small voids while away from the spall plane the material experiences a uniform compressive shock deformation.

2. Experimental

The x-ray synchrotron measurements were made with a 3D X-ray crystal microscope[6] based on polychromatic (white) beam with the energy range 6 - 25 keV. This method allows for true 3D mapping of the crystalline phase, orientation and plastic deformation with unprecedented spatial resolution. A region of interest is identified using an optical microscope and the polychromatic x-ray beam is focused onto the sample. The crystallographic orientation at each position of the sample is precisely determined by an automated indexing program. Polychromatic microdiffraction produces misoriented overlapping Laue images from each deformation cell intercepted by the x-ray beam. The challenge is to extract the local orientation along the incident beam. The process of disentangling the Laue pattern to determine the local orientation can be carried out either by a direct experimental technique called differential aperture microscopy[6] or by pattern matching/ absorption sensitive methods. Once the Laue pattern from a subgrain volume has been obtained and the indices of each reflection determined, powerful single-crystal methods can be employed to determine the unit cell parameters. Although differential aperture microscopy (DAXM) can often resolve Laue image into single crystal-like patterns, when the density of unpaired or “geometrically necessary” dislocations (GNDs) is high, streaking is observed even for microsize regions in DAXM resolved patterns.[7 - 9] Typically streaking in Laue patterns indicates the presence of lattice rotations within the probed region as a result of the GNDs formation. Data collection has been carried out using microbeam Laue diffraction on beamline 34ID at the Advanced Photon Source. Dimensions of the beam were 0.5 by 0.5 microns with a penetration depth of ~ 500 micron. Details on the experimental setting and data collection can be found elsewhere.[6-9]

A blown up sketch of the shocked Al and Cu single crystals and the geometry of initial and diffracted white beam radiation and the microbeam geometry are shown in the Fig. 1. An Al (123) single crystal with initial dimensions of 5 mm in diameter by 5 mm in thickness was cut along the shock direction and analyzed at different locations of the sample cross section (Fig 1b). A Cu (001) single crystal oriented for multiple slip with initial 3 mm by 1 mm thickness was analyzed from both the back side and the crater (Fig. 1c).

3. Results and Discussion

A number of experimental studies of plastic deformation under different loading conditions show that cell-wall structures with alternating regions of high and low dislocations density are typically formed.[8 - 13] The dislocation walls contain a high density of dislocations, and cell interiors have a very low dislocation population. Shock often causes the formation of a high density of voids in materials.[4]

Refs. [7 - 9] describe an approach to characterize the dislocation structure and determine the activated slip systems by analyzing the streaked Laue pattern using a multiscale hierarchical framework. In white beam diffraction the full width at half maximum of the streak (ξ direction) $FWHM_{\xi} \equiv \delta m_{\xi}$ is a function of the misorientation within the probed region caused by geometrically necessary dislocations (GNDs). The orientation of the GNDs influences the character of Laue spot. In the transverse direction ν, the $FWHM_{\nu} \equiv \delta m_{\nu}$ depends on the total number of all dislocations (GNDs and statistically stored, SS) per unit length and usually $\delta m_{\xi} >> \delta m_{\nu}$.

Under multiple slip the Laue intensity distribution depends on the dislocation density tensor ρ_{ij}. The element of dislocation density tensor can be written as:

$$\rho_{ik} = \tau_i b_k \delta(r). \qquad (1)$$

The magnitude of the vector, $\boldsymbol{\tau}$, is the net number of dislocations having Burgers vector, $\boldsymbol{b}$, crossing unit area normal to $\boldsymbol{\tau}$.[14] Following the approach[15] in the framework of strain gradient plasticity, the total GNDs and GNBs density tensor ρ_{ij} relates to the strain gradient tensor η_{lmk};

$$\varepsilon_{ilm}\eta_{lmk} = -\rho_{ik}; \quad \eta_{lmk} = \partial^2 u_k / \partial x_l \partial x_m \qquad (2)$$

Here ε_{ilm} is the anti-symmetric Levi-Civita tensor. Under multiple slip, intensity distribution around each reciprocal lattice point and the streak direction ξ of the Laue spot depends on the aforementioned dislocation density tensor. Here u_k, the k-th component of the displacement field $\boldsymbol{u}$ for any unit cell, is due to all dislocations in the crystal.

We calculate the total displacement of the i-th cell $\boldsymbol{u}_i$ from the equilibrium positions $\mathbf{R}_i^0$ corresponding to the undeformed crystal using continuum elastic theory. This displacement is due to all dislocations and is defined using random numbers c_t by the equation[6 - 8]:

$$\mathbf{u}_i = \sum_t c_t \mathbf{u}_{it} \; ; \qquad c_t = \begin{cases} 1, (+) \\ 0, (-) \end{cases} \tag{3}$$

If there is a dislocation at position $t, c_t = 1$, and for positions without dislocations $c_t = 0$. Dislocation density tensor ρ_{ik} gives the sum of the Burgers vectors of the dislocations whose Burgers vector and line direction are directed parallel to x_k and x_i axes (k, i =1, 2, 3), respectively. These are set as an initial input parameter for simulation of the Laue pattern. By simulating Laue images with different GND slip systems, we can determine the set of slip systems that are best fit to the local lattice curvature and experimentally determined Laue image. The analysis of the dislocation substructure for multiple slip deformation includes the following steps:

- Determination of the local lattice curvature (misorientation axes and angle) from the experimental Laue pattern at each probed location.
- Performing least square fit find the components of the dislocation density tensor.

3.1 Inhomogeneous plastic deformation in the shock recovered Al single crystal

Polychromatic x-ray microdiffraction (PXM) technique was applied to the complicated dislocation structure arising from the shock induced plastic/elastic deformation of the Al (123) single crystal. The microbeam-Laue diffraction revealed several distinct zones located at different depth under the shock front (indicated by green arrow in Fig.1 b). Pronounced streaking of Laue images is observed in the zones 1 and 7 close to the front and back surface of the after shock (123) Al single crystal (Fig. 2).

Much lower GND density is observed in the zones 2, 3, 4 located approximately in the upper half of the sample and zone 6 close to the bottom (Fig. 3a). A high density of GNDs is observed in the near surface zones both near the crater (zone 1) and near the back surface (zone 7). The most interesting images correspond to the zone 5 (Fig. 2 e, Fig. 3a). Distance – time plot (Fig. 3 b) obtained similar to the Ref. [4] shows that the impact of a projectile of thickness l_p on a target of thickness $2l_p$ results in the formation of tensile region, and the position of the tensile stress maximum exactly coincides with the maximal GNDs density distribution found in this sample. This zone corresponds to the region in which X-ray tomography, SEM and OIM indicate intensive void formation (Fig. 4). This zone not only has a high density of GNDs, but the crystallographic orientation alternates in this zone.

Experimental and simulated Laue patterns obtained from the near-surface crater region (zone 1) are shown at Fig. 5 a, b. The aforementioned analysis indicates that activated slip systems in this region result in lattice rotations around an axis ~ perpendicular to the shock direction. Asymmetric intensity profiles observed for all Laue streaks indicate the gradient of GNDs density with depth in the near surface region of the shock recovered Al single crystal.

3.2 3D Spatially resolved measurements

To get a better understanding of the reasons for such a peculiar shape of the Laue image 3D depth resolved measurements[6] were performed. This differential aperture

measurements track changes in the Laue pattern at different depth along the beam. They show that in this region alternating local lattice rotations take place (Fig. 6). We argue that such alternating local rotations are caused by the presence of voids creating regions of plastic deformation in the vicinity of the void. This result agrees well with SEM and OIM observations.

The spallation fracture is created by the shock waves and their release from the free surfaces. In the gas gun experiment, tension first occurs when the release wave from the front surface and the release wave from the impact side meet.[2 - 4] The experiment is designed so that this geometric point (defined by the wave speeds) is approximately centered in the sample. One often speaks of a "spall plane", however this is never true for a real material. There are several modifying effects: (1) the lowest tension void nucleation sites are the inclusions, which are not distributed on a plane; and (2) the stress waves travel faster than the rate of void growth, which means that regions away from the "spall plane" reach the nucleation threshold before the region at the spall plane completes its stress relaxation. The damage distribution is sharper than a Gaussian and looks more like a centered exponential or algebraic form. For the Al experiments with impacts for ~200m/s and sample targets of 4-5mm, the FWHM of the damage zone was consistently about microns (0.4mm).

3.3 Inhomogeneous plastic deformation in the shock recovered Cu single crystal

2D map of PXM intensity was collected from different locations at the back and front sides of the Cu single crystal with a step size of 1 μm in both directions. Typical Laue patterns from the back and front sides of the shocked Cu crystal are shown in (Fig. 7). One of the Laue pattern (Fig. 7 a) is indexed for convenience. Residual plastic deformation is observed at both sides of the crystal. However deformation at the crater is much more severe. Formation of geometrically necessary dislocations (GNDs) and geometrically necessary boundaries (GNBs) is observed.

As was shown previously GNDs and GNBs cause streaking and splitting of the Laue spots [7 - 9]. The direction of the streak is related to the dislocation density tensor in the probed region and to the local lattice rotations. Figure 8 shows fluctuations in the direction and intensity distribution of (006) Laue spot at different locations at the back side of the (001) Cu single crystal.

An inhomogeneous distribution of GN dislocations and GN boundaries is observed on both the front and back sides of the shock recovered Cu single crystal. GNDs are grouped within mesoscale regions of large lattice rotations (red color in Fig. 9).

Severe plastic deformation in the crater is accompanied by the formation of a large number of twins. Twinning causes splitting of all Laue spots and appearance of “two” overlapping disoriented diffraction patterns (Figs. 7 d, e). This is in agreement with TEM results described in the Ref. [17].

The formation of narrow slip bands separating regions with low GN density can be understood from the view point of mesoscopic structure formation: The free energy of the system decreases due to the formation of mesoscopic self organized structure. As a result of multiple slip and fragmentation rough boundaries are formed. With increasing strain due to collective behavior in the interacting dislocation ensemble, rotational plastic

modes appear. Rotations are localized in the narrow microbands separating the crystal into strongly disoriented regions. This evolution can be schematically described in the following structural steps: (1) randomly distributed dislocations, (2) cell-wall structure; (3) microbands with IDBs and GNBs. With an increase of deformation the cell boundaries become narrower and result in the formation of IDBs and GNBs. At the micro and meso scales the spontaneous reconstruction of the dislocation substructure is a typical sign of the nonlinear behavior of the deformed material. The statistically stored (SS) dislocation density is the most important parameter at the low strain deformation without strain gradients. With increasing strain SS randomly distributed dislocations reconstruct with a formation of the cell-wall structure.[10, 18] During and after shock strain gradients and the density of GNDs and GNBs control formation of microbands with localized rotational modes. Formation of each type of dislocation substructure depends not only on the shock loading parameters but also on the interaction between already existing dislocations (and defects) resulting in the collective phenomena in the dislocation ensemble.[19]

4. Conclusions

Microbeam-Laue diffraction reveals several distinct zones located at different depths under the shock front. Laue images demonstrate pronounced lattice rotations in the zones close to the front and back surface of the after shock (123) Al single crystal. The formation of geometrically necessary dislocations in these zones is consistent with a single slip mode in the near surface regions. In the interior of the Al single crystal the portion of geometrically necessary dislocations in the dislocation ensemble decreases, and the portion of statistically stored dislocations increases. The ratio between GN and SS dislocations reverses in the spallation region. An intensive mesoscale void formation is observed in the region close to the center of the samples, which is accompanied by a complicated shape of the Laue diffraction peaks. The density and organization of dislocations is presented as a function of depth under the shock front as well as comparison to SEM on the same samples. In the shock-recovered Cu single crystal mesoscale inhomogeneities in GN dislocations distribution are observed both at the crater and the back sides.

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Figure Captions

Fig. 1. 3D polychromatic microbeam geometry: wire (yellow circle) is shadowing different portions of the diffracted intensity on a CCD (a); blown up sketch of the Al (b) and Cu (c) single crystals orientation during shock experiment and orientation relative to microbeam.

Fig. 2. Laue images obtained at different depth under the shock front of (123) Al single crystal: a) zone 1 near the shock front; b) zone 2; c) zone 3; d) zone 4; e)zone 5 corresponding to the region of intensive void formation; f) zone 7 near the back surface of the shocked single crystal.

Fig. 3 (a) GNDs density oscillations with the depth of the shock recovered Al single crystal; (b) a distance – time plot of the shock wave propagation after high-velocity impact according to the Ref. [20].

Fig. 4. (a) X-ray tomography shows region with high concentration of mesoscale size voids in the spall region; (b) SEM image of the void at the surface of the cross section of the after shock Al single crystal chosen for PXM analysis; (c) OIM image of the orientation change around the same void, and color scale of orientation distributions (d).

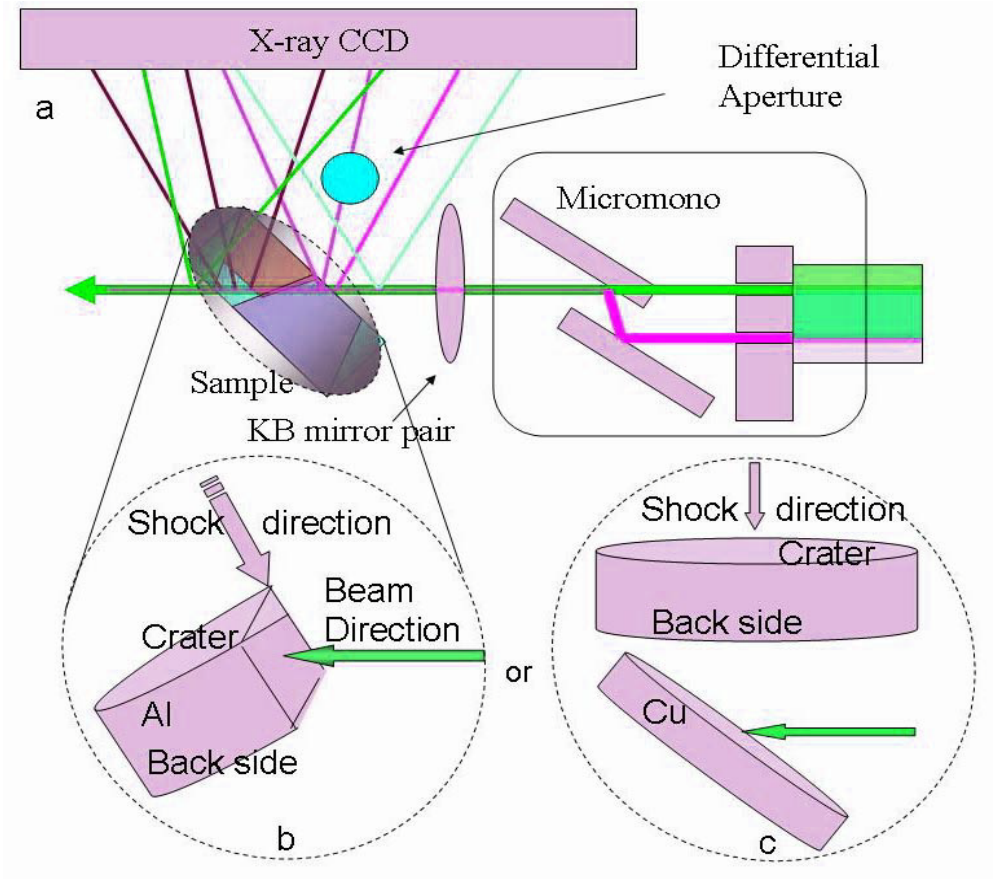
Fig. 5 Experimental (a) and simulated (b) images from a near surface crater region (zone 1) of the shock recovered Al single crystal are consistent with a “macroscopically” single slip deformation.

Fig. 6. Total (left top corner) and depth resolved Laue patterns from the spall region with voids. Depth is shown in microns at each depth resolved images. Total Laue pattern is indexed for convenience.

Fig. 7. Laue images obtained from the (001) Cu shocked single crystal: The back side of crystal (a - c) and the crater at the front side of the crystal (d – f).

Fig.8 (006) Laue spot at different locations of the back of the shocked (001) Cu single crystal.

Fig. 9. Inhomogeneous GNDs distribution in the shock recovered Cu sample: (a) the crater region; (b) the back of the sample.



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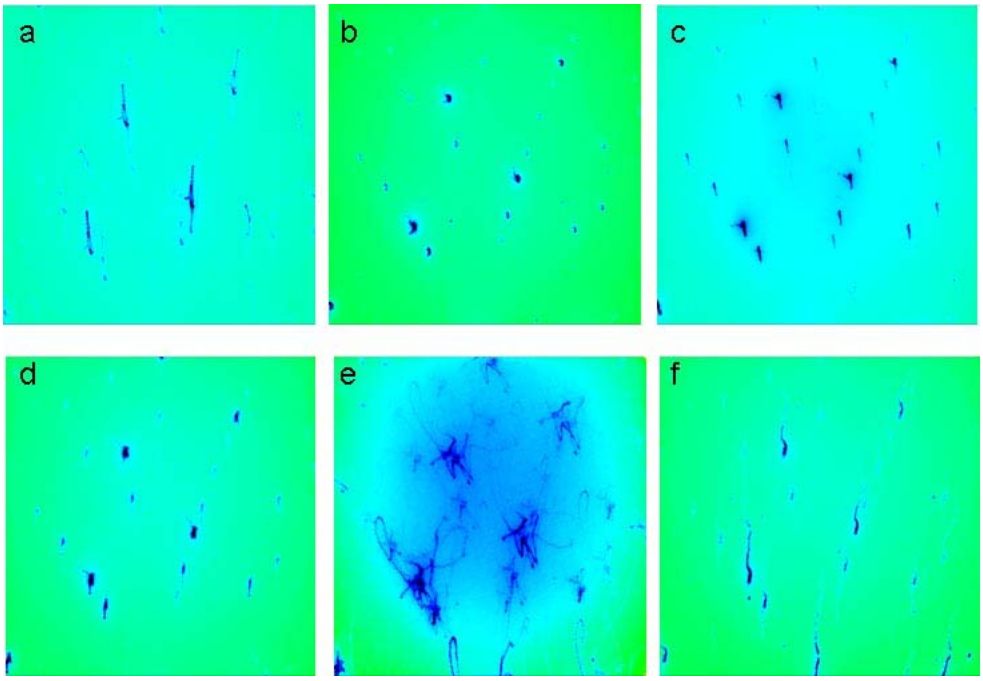

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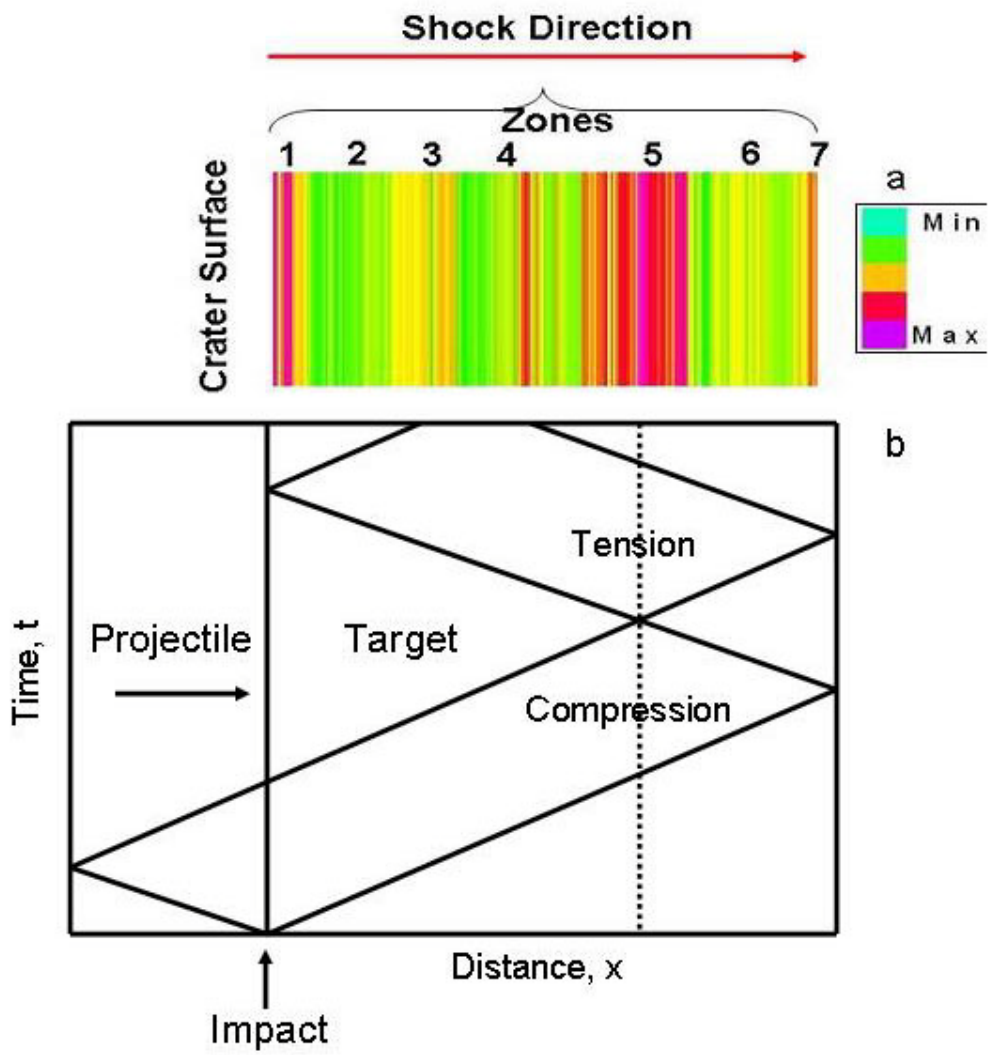

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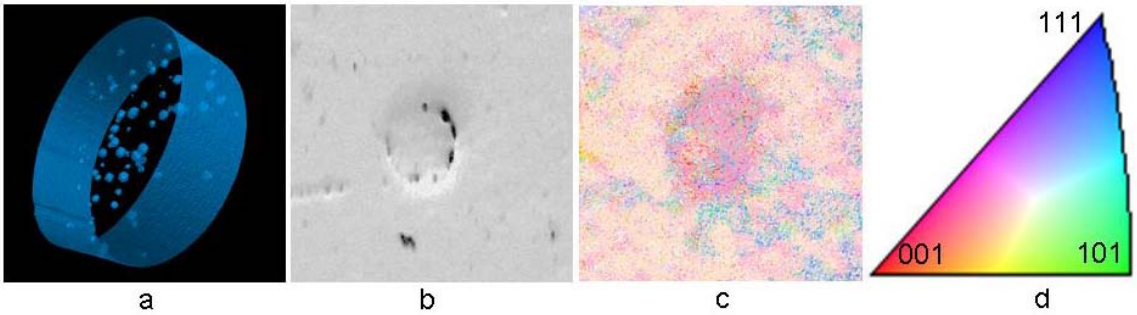

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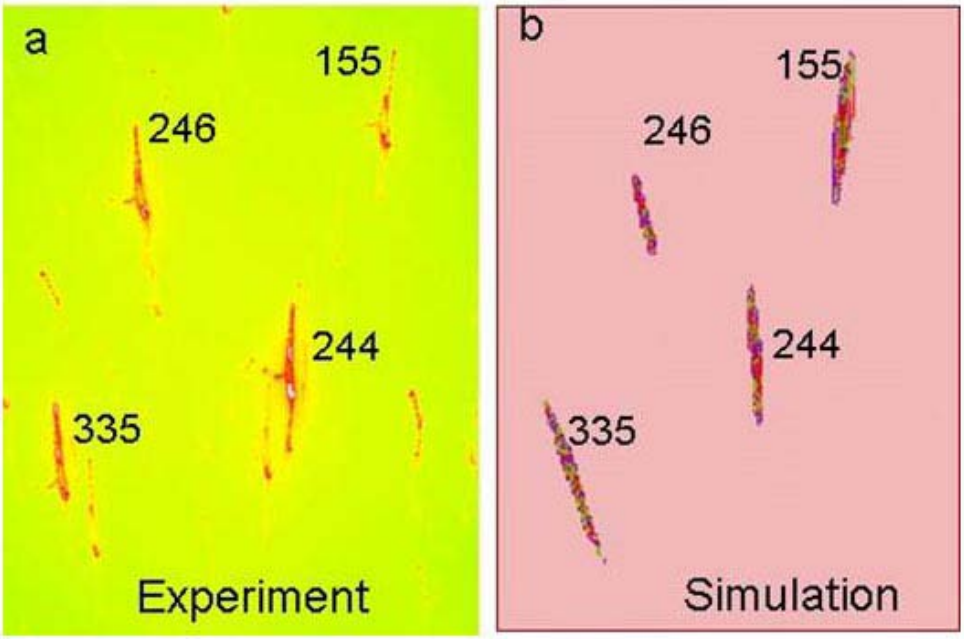

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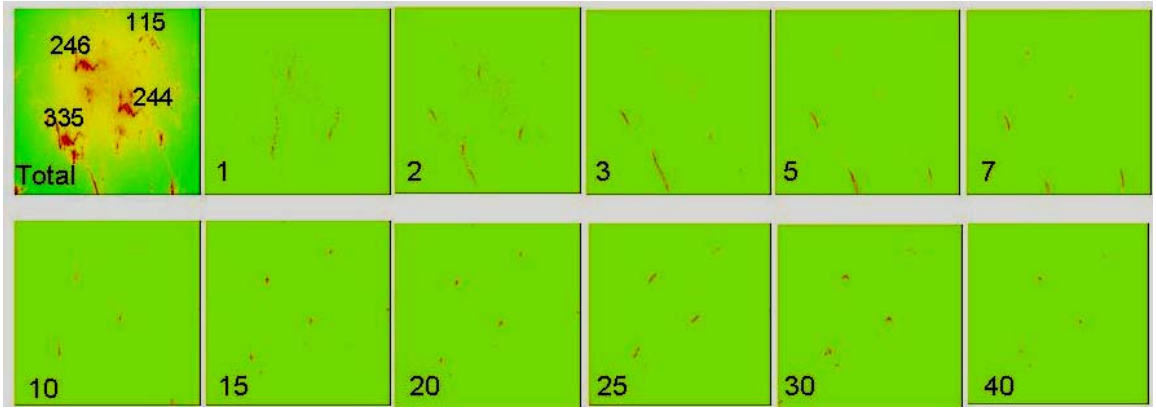

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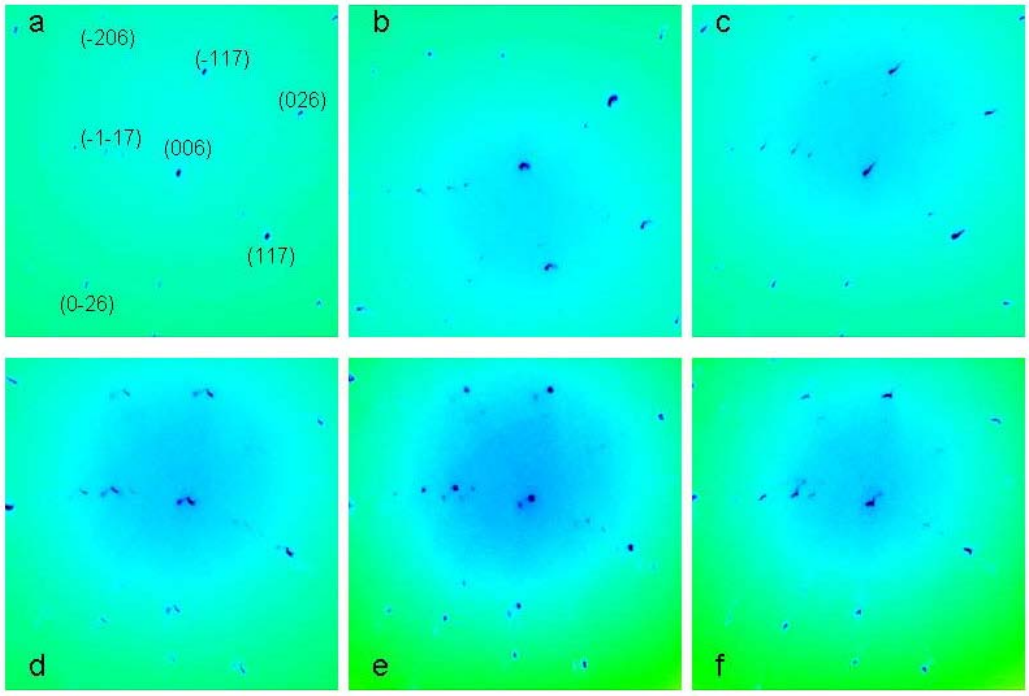

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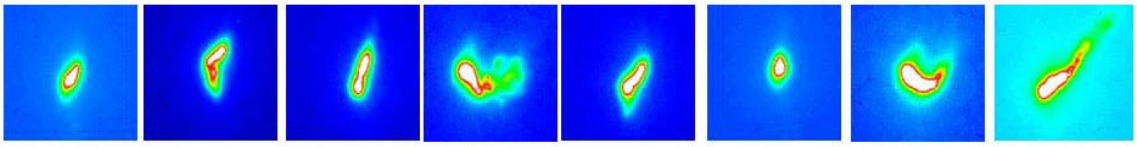

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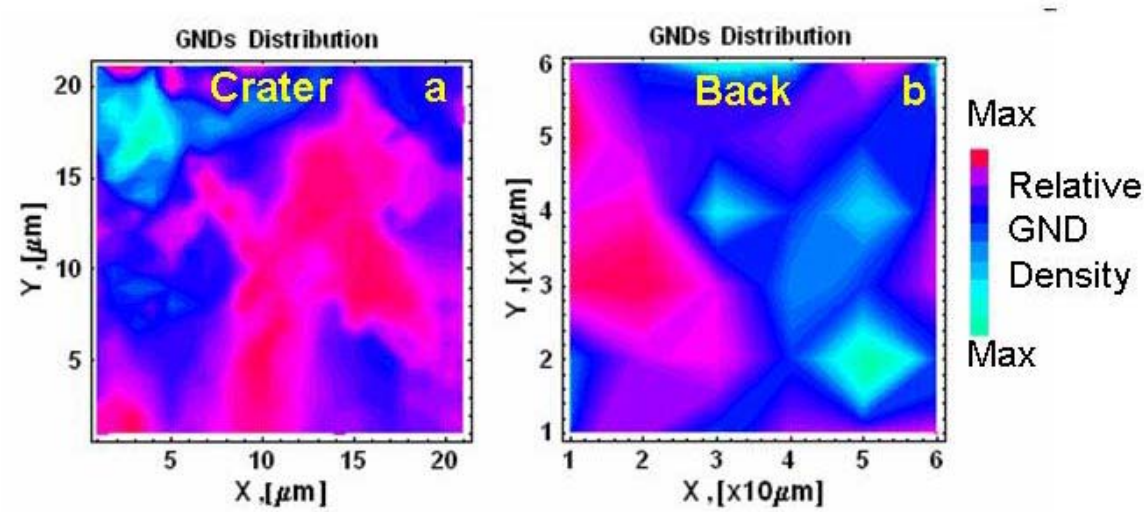

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