

## THE 3D X-RAY CRYSTAL MICROSCOPE: AN UNPRECEDENTED TOOL FOR ICME

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### Abstract

There is a long-standing debate over the length scales needed to understand the behavior of materials and the role of surfaces, defects, and inhomogeneities. Indeed the properties of most materials are ultimately determined by defects –including grain boundaries and surfaces—that are either introduced during processing or in-service, and defect density and distribution must be considered for high-fidelity integrated computational modeling and engineering. Scientists at ORNL together with partners at Argonne have developed a powerful 3D X-ray Crystal Microscope that can nondestructively characterize the local 3D crystal structure of polycrystalline materials with submicron resolution and with sensitivity to the local crystal structure, orientation, elastic strain tensor and the local Nye tensor. This emerging tool provides unprecedented tests of materials models under different processing/environmental conditions and provides new insights into the impact of unpaired dislocations, elastic strain and surfaces and interfaces. The promise of the 3D Microscope and the emergence of similar instruments at synchrotrons around the world will be discussed with respect to ICME.

### Introduction

Although the emphasis in Integrated Computational Materials Engineering is on *Integration* and *Engineering*, the underlying complexity of materials modeling is a grand challenge that prevents widespread adoption of ICME [1]. To compensate, data-base methods can be used to extend our understanding of materials properties, but ultimately transformational products will depend on the ability to predict structure property relationships arising from processing and in-service. Indeed underlying ICME is a need to understand materials defects on all length scales.

The mesoscale- where the collective interactions of defects are difficult to model with atomic resolution and where the three-dimensional (3D) behavior of materials is difficult to characterize experimentally- is particularly challenging. This is a scale too large for atomic models, and often too deep for atomic characterization with electron microscopy. Here we describe the 3D X-ray Crystal Microscope [2,3] and how it can provide unprecedented information about local crystal structure and defect organization with mesoscale resolution. Because x-ray characterization is

inherently nondestructive for many materials, the 3D characterization of local crystal structure can be used to test existing models and to guide the development of new models, which incorporate defect physics at a new level.

The 3D X-ray crystal microscope is based on Laue diffraction with the ability to accurately measure the energy of each Laue spot as needed [2]. Because a polychromatic beam is essential for Laue diffraction, achromatic focusing optics are essential and the 3D X-ray Crystal Microscope is a specialized version of polychromatic microdiffraction with the ability to resolve the structure of volumes both transverse to the beam and along the penetrating polychromatic x-ray beam. With Laue diffraction, 4 linearly independent reflections are sufficient to determine the unit cell shape and orientation and if the energy of at least one reflection is determined, then the unit cell volume is also measured [2]. In practice, even simple crystal structures reflect 8-20 Laue spots for x-ray bandpasses from 12-18 keV. Precision measurements of local unit cell parameters is made possible by the use of submicron focused x-ray beams, with highly accurate x-ray sensitive area detectors. The general scheme for a 3D X-ray crystal microscope is shown in Fig. 1. As illustrated, X-rays from a brilliant synchrotron source are focused to a submicron spot using wide-bandpass total-external-reflection mirror optics. Laue patterns generated by the penetrating beam, are measured by an area detector set normal to the incident beam. The overlapping patterns from each subgrain illuminated volume element along the beam are disentangled by passing an absorbing 20-80  $\mu\text{m}$  diameter wire near the sample surface.<sup>3</sup> As the wire moves, changes in the diffraction pattern are due to rays that pass near the front or back surface of the wire [3]. By ray tracing from each pixel on the area detector, past the edge of the wire and onto the incident beam, the individual Laue patterns are determined for subgrain volumes along the beam; because the wire is much closer to the sample than to the detector, a 25-100  $\mu\text{m}$  displacement on the detector has a submicron effect on the ray traced origin of the scattering.

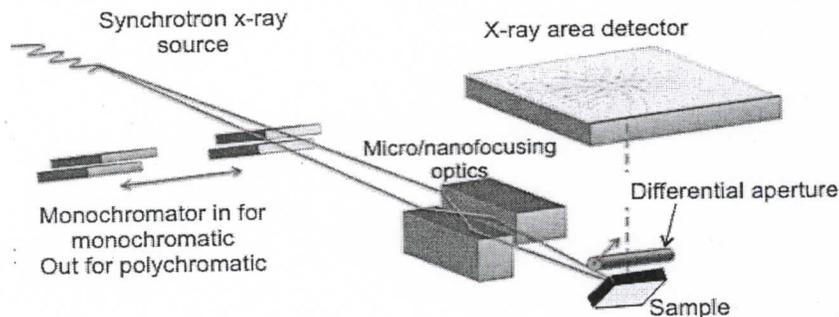
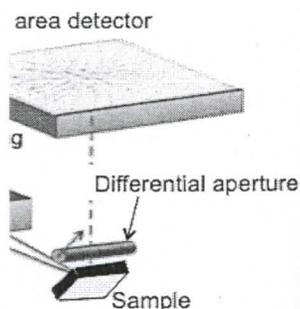


Fig. 1. Key components of a 3D X-ray crystal microscope. The ultra-brilliant x-ray source produces a polychromatic beam that is focused by nondispersive elliptical x-ray supermirrors. The beam can either be passed onto the mirrors as a monochromatic or a polychromatic beam. Diffraction from the sample is detected by an area detector and the signal from along the penetrating beam is resolved using the differential aperture, which passes close to the sample surface.

Although the method sounds simple, there is a considerable sophistication in all aspects of the instrumentation and software. Precision elliptical x-ray super mirrors and alignment hardware are needed to focus the incident beam to 200-500 nm [4,5]. Special stages are needed that can work

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with tens of nanometer accuracy in a "fly scan" mode for rapid data collection. Sophisticated data handling is required to process the ~1 billion bits/sec generated at a 10 frame/sec data collection rate. Over a 24 hour period, this adds up to around 80 terabits ( $80 \times 10^{12}$ ). In addition, a specially designed monochromator is used to precisely determine the energy of Laue spots or to obtain full reciprocal space volumes from samples [6]. This monochromator must have an absolute energy calibration and must have the ability to sweep wide energies and be inserted into the beam or withdrawn as needed. Below we describe some example applications of 3D X-ray Crystal microscopy and how they impact ICME.

## Applications

### Heterogeneous Deformation in Polycrystalline Materials

It is well known that polycrystalline materials deform in a heterogeneous manner. Deformation inhomogeneities arise from crystallographic orientation anisotropies, from discontinuities set by surfaces/interfaces and from inhomogeneous defect distributions or boundary conditions. Although most previous work has concentrated on ensemble average behaviors in polycrystalline materials, the development of the 3D X-ray crystal microscope now allows for a nondestructive evaluation of local crystal structure *before and after* deformation. This allows for detailed studies of the local conditions around a subgrain sample volume and how the local conditions influence deformation. Questions of interest include, how do surfaces affect deformation? How does grain boundary type influence the constitutive equations and how do surrounding grain orientations influence local deformations?

Ensemble-average strain measurements in polycrystalline materials have identified significant differences in the average strain of different phases or of grains with different crystallographic directions with respect to the macroscopic strain axis. These differences are understood in terms of compliance of the heterogeneous materials or anisotropic grains [7,8]. More recently, high-energy spatially-resolved local crystal structure measurements at the ESRF provided a first test of theories describing heterogeneous *rotations* of grains depending on their crystallographic orientation with respect to the macroscopic deformation direction [9]. These experiments identified both a strong tendency to follow the rotation direction predicted by the original grain orientation, and some anomalous behaviors. Two experiments led by Pang and co-workers now provide tantalizing insights into the fundamentally important role of surfaces and interfaces in heterogeneous polycrystalline deformation.

In one experiment [10], the local orientations of crystal volumes near grain boundaries were measured. The sample was then deformed and re-measured. The *changes* in crystal orientation across the grain boundaries were plotted against the original difference in orientation. As might be expected, there is a roughly linear relationship between the grain boundary misorientation before deformation and the *change* in misorientation after deformation. However, for some coincident site lattices, there are strong deviations from the overall trend, with a wide distribution in misorientation change including some cases with almost no change (Fig. 2 left). This points to the important role of grain boundary type in local deformation. Moreover, measurements of many deformed polycrystalline samples find a general trend for much larger curvatures in the crystallographic lattice near grain boundaries and surfaces than in grain cores. It appears that the core of individual grains tend to rotate as a unit, while the surface accommodates the rotation with respect to the surrounding grains.

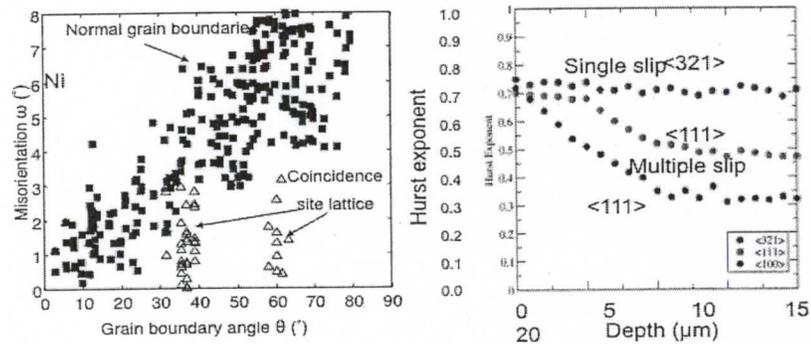


Fig 2. Left. The change in misorientation across a grain boundary follows a roughly linear relationship except near coincidence site lattices. Right. The Hurst coefficient correlating rotations near the surface of a deformed crystal has a universal behavior independent of crystal orientation at the surface, but is dependent on crystal orientation deeper into the sample.

An even more interesting set of experiments has looked at the deformation structure of a single crystal near its surface. Previous experiments with surface roughness found that roughness after deformation had a self-affine nature with a universal Hurst coefficient independent of crystallographic orientation [11]. A question of interest, was whether such a universal behavior also applied to crystallographic rotations, and if the nature of the rotations would change below the free surface. Pang and co-workers found that rotations had the same universal Hurst coefficient as roughness at the surface of crystals, independent of crystallographic orientation, but that below the surface, the Hurst coefficient changed (Fig 2 right) [12]. The depth below the surface where the Hurst coefficient changed depended on the number of slip systems activated by the deformation geometry. These results point to the importance of free surfaces, grain orientation and grain boundaries in local deformation with important implications for the constitutive equations at grain boundaries and surfaces.

#### Nye Tensor

The Nye Tensor is a fundamental variable in finite plasticity models, yet measurements of the full Nye tensor have not previously been possible; the Nye Tensor depends on curvature, and elastic strain in a 3D volume. Now the 3D X-ray Crystal Microscope allows for precision nondestructive measurements of local crystal curvature and the elastic strain tensor in three dimensions; this information is required for a direct comparison to finite plasticity models at a local (submicron) level. Because local curvature can arise both from unpaired dislocations, and from elastic strain gradients a simple measurement of lattice curvature is insufficient, as illustrated in a series of experiments by Larson and co-workers [13]. In these measurements single crystals of Si were bent above and below the elastic to plastic transition temperature. In measurements below the transition, maps of the Nye tensor are featureless and show structure at the uncertainty of the measurements whereas maps of the elastic strain tensor follow the anticipated patterns from bent beam mechanics (Fig 3a). For crystals bent above the elastic to plastic transition temperature the Nye Tensor maps show a strong spatial dependence whereas

the elastic strain tensor maps are featureless (Fig 3b). This proof-of-principle experiment opens up the possibility of direct verification of plasticity models for inclusion into ICME.

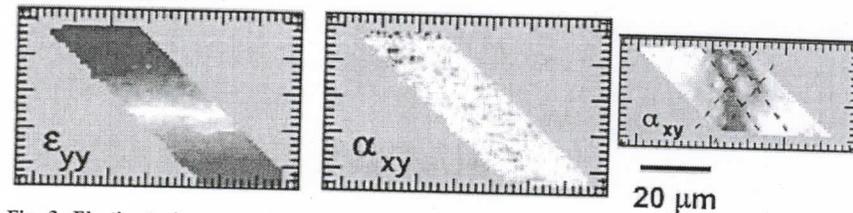


Fig. 3. Elastic strain tensor element (right) for elastically bent silicon beam, dislocation tensor middle for the same elastically bent silicon beam, and dislocation tensor from a plastically bent silicon beam [ref. 13]. The full scale color is  $\pm 3 \times 10^{-3}$  for the elastic strain, and  $3 \text{ mrad}/\mu\text{m}$  for the middle and right images.

#### Grain growth

Thermal treatment after deformation is a classic tool for controlling the microstructure of materials and as such is an essential element that must be clearly understood for ICME modeling of materials microstructure/processing behavior. Current models of grain growth are dominated by measurements of surface grain structures, which have inherent assumptions, or by serial sectioning which provides ensemble-average information about grain sizes and distributions. However, detailed nondestructive volumetric maps before and after grain growth are just now becoming a reality. Such maps can be obtained using high-energy monochromatic three and four-dimensional (time) measurements [14] or with the 3D X-ray crystal microscope. Although the 3D X-ray crystal microscope is considerably slower in collecting data, it provides submicron resolution, and simultaneously quantifies the unpaired dislocation density. This information is unprecedented and provides a direct measurement of the grain boundary motions and grain refinement to test models. Early measurements by Budai indicate that the defect density of grains plays an important role in future grain boundary motion [15]. Future measurements will guide and test models of grain growth with unprecedented fidelity.

#### Summary and Resources Around the World

X-ray microscopy is an emerging field, which provides new insights into materials behavior [16]. The promise of x-ray microscopy in general, and polychromatic methods in particular is driving the development of new instrumentation around the world. The principles for the 3D X-ray Crystal Microscope were first developed on beamline 7 at the Advanced Photon Source (APS), and then transferred to a permanent station 34-ID-E at the APS. About the same time, a two-dimensional polychromatic microprobe was installed at the Advanced Light Source, and it has since been upgraded with a higher brightness super bend source and with the hardware for 3D imaging. Other polychromatic beamlines exist at the Canadian Light Source, the Pohang Light Source and the Swiss Light Source, with other development programs at the European Synchrotron Radiation Facility, Spring 8 and elsewhere. Additional beamlines are under development at the Australian Light Source and the Taiwan Light source. There are also rapid developments in high-energy monochromatic techniques, which seek to achieve 3D submicron resolution grain mapping with much faster data collection. This new instrumentation and technique

development will provide unprecedented opportunities for guiding and testing mesoscale modes with direct implications for ICME.

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