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# Three-Dimensional Micron-Resolution X-Ray Laue Diffraction Measurement of Thermal Grain-Evolution in Aluminum

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**Abstract.** A new technique for investigating 3D grain growth in polycrystalline materials using white x-ray microdiffraction with micron point-to-point spatial resolution is presented. This technique utilizes focused polychromatic x-rays at the Advanced Photon Source, differential aperture depth-profiling, CCD measurements, and automated analysis of spatially-resolved Laue patterns to measure local lattice structure and orientation. 3D thermal grain growth studies of hot-rolled aluminum have been initiated to demonstrate the capabilities of this method. Complete 3D grain orientation maps were obtained from a hot-rolled aluminum polycrystal. The sample was then annealed to induce grain growth, cooled to room temperature, and re-mapped to measure the thermal migration of all grain boundaries within the same volume region. Initial observations reveal significant grain growth above 360ºC, involving movement of both low- and high-angle boundaries. Systematic measurements have been obtained of the as-rolled grain structure and of the microstructural evolution after annealing at successively higher temperatures. Small second-phase precipitates have been identified. Such measurements will provide the detailed 3D experimental link needed for testing theories and computer models of 3D grain growth in bulk materials.

## Introduction

The importance of understanding three-dimensional (3D) grain growth for controlling materials properties is well-recognized, and a large number of experimental and theoretical studies have investigated fundamental microstructural mechanisms associated with thermal processing. In particular, large computer simulations have become increasingly important in modeling recrystallization and growth processes at the mesoscopic level with increasingly realistic microstructures. Experimentally, penetrating probes such as neutron or x-ray diffraction have typically been used to provide measurements of texture, average grain sizes, and strain in bulk volumes. In addition, surface probes such as optical microscopy or electron backscatter diffraction [1-3] have been combined with serial sectioning to provide 3D spatial characterization of grain morphologies at particular stages of materials processing. More recently, advances in 3D x-ray microscopy are now enabling non-destructive, in-situ measurements with high spatial resolution in bulk polycrystalline samples. These advances have been made possible by the availability of high-brilliance synchrotron x-ray sources, the development of digital area detectors and large improvements in precision x-ray focusing optics [4]. Consequently, research efforts applying various x-ray microscopy techniques to study microstructural evolution during materials processing have been initiated at several synchrotron facilities [5-7]. At the European Synchrotron Radiation Facility (ESRF), a 3D X-Ray Diffraction (3DXRD) microscope uses monochromatic, penetrating

(E >50 keV) x-rays and a tomographic approach to obtain lattice orientation and strain with ~5 μm spatial resolution [8]. Examples of 3DXRD investigations include nucleation and growth studies of pearlite and ferrite grains during the cooling of steel [9], and studies of recrystallization kinetics for individual grains in cold-rolled aluminum [10]. In the present paper, complementary techniques using polychromatic (Laue) x-ray microdiffraction being developed at the Advanced Photon Source (APS) are presented [11-12]. In our approach, lower-energy (~8–23 keV) synchrotron radiation is focused to submicron diameter, and scanning mode operation yields the full Laue diffraction pattern from each submicron volume element within a 3D region. Initial microdiffraction investigations have revealed detailed information describing local deformation in plane-strained aluminum [13] and in microindented copper crystals [14]. In this paper, the polychromatic 3D technique will be briefly described and its potential for elucidating 3D grain growth will be demonstrated using initial results from in-situ measurements of the evolution of particular grain boundaries during thermal annealing of polycrystalline aluminum. This type of information in 3D is precisely the experimental guidance currently needed for theoretical and modeling treatments of mesoscopic grain growth in bulk materials. The long-term goal of these studies is to combine nondestructive 3D microstructural measurements with large-scale computer modeling efforts in order to understand grain dynamics under particular processing conditions [15,16].

**Experimental**

Fig. 1 shows schematically the experimental setup located at the UNICAT sector 34 beamline at the APS. Synchrotron radiation shown on the right is intentionally taken 50 μrad off-axis from an undulator device in order to obtain a more uniform polychromatic (white) spectrum. The white beam is incident at a glancing angle on a crossed pair of high-precision, elliptically-figured Kirkpatrick-Baez (K-B) mirrors. The K-B mirrors non-dispersively focus the white beam to a diameter of <1 μm near the sample position.

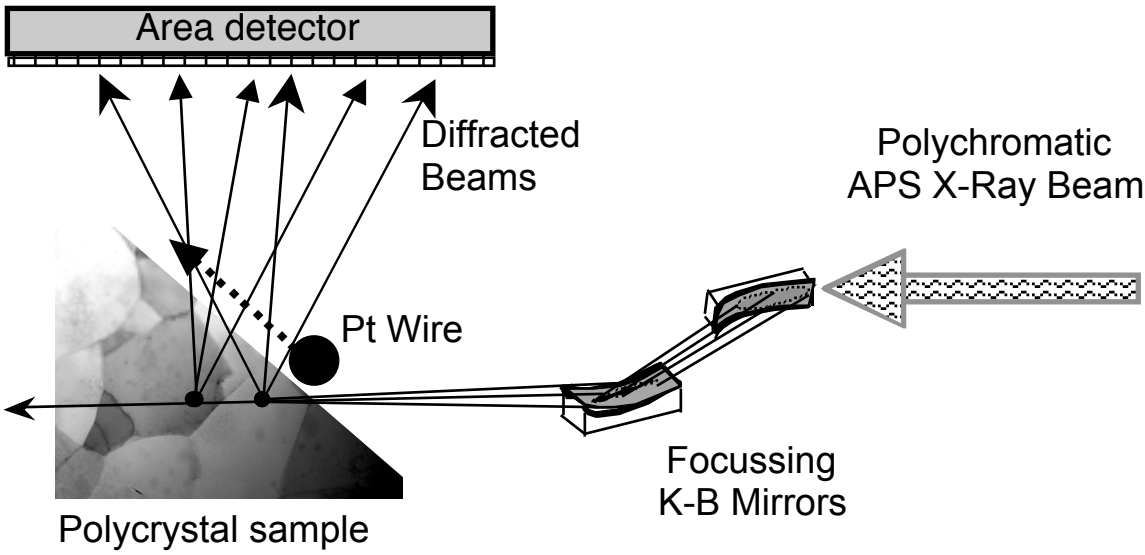


**Fig 1**. Schematic diagram illustrating components of 3D polychromatic x-ray microdiffraction. Synchrotron radiation from an APS undulator is incident from the right and is focused to ~0.5 μm diameter onto a polycrystalline sample by a crossed pair of K-B mirrors. Laue diffracted beams are measured by the CCD area detector and their point of origin along the incident beam is determined by depth-profiling using the Pt wire.

As the microbeam passes through a particular grain in a polycrystalline sample, Bragg's Law determines the energies and directions of the diffracted beams and a full Laue diffraction pattern (many Bragg peaks) is generated from each individual irradiated grain. In this case, the samples were polycrystalline slices taken from a thicker slab of 1xxx series aluminum (~1% Fe,Si) hot-rolled at 200°C and mounted on a small in-situ heating stage which is, in turn, mounted on a high-precision translating stage. Note that the use of white radiation means that no sample rotations are needed to obtain diffraction patterns. The Laue patterns are collected digitally using a charge coupled device (CCD) area detector. Since the incident beam irradiates a large number of grains with different orientations as it penetrates the sample, many Laue patterns are superimposed in a single raw image (e.g. Fig. 2a).

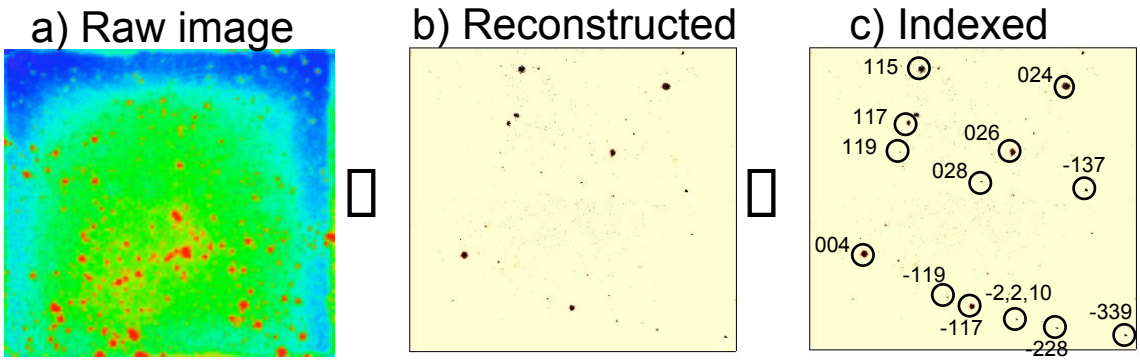


Fig 2. Typical CCD diffraction images from different stages of data analysis: a) Raw diffraction image consisting of superimposed Laue patterns from all grains irradiated by the incident microbeam; b) Reconstructed image from a particular ~1 $\mu m^3$ volume element; and c) Indexation of reconstructed image reveals local lattice orientation.

In order to depth resolve patterns from different grains or from different positions within a single large grain, a differential aperture x-ray microscopy (DAXM) technique has been developed [11]. In a manner analogous to a step-scanned pinhole camera, a series of raw diffraction images are taken as a strongly absorbing 50 μm diameter Pt wire is translated in ~ 1 μm steps parallel to the sample surface (Fig. 1). Data analysis based on subtracting images from different wire positions uniquely determines the point of origin along the incident beam for all diffracted intensities. The result of the analysis is a series of reconstructed Laue images with each pattern containing the Bragg scattering diffracted from a particular ~1 $\mu m^3$ sample region irradiated by the incident microbeam. As illustrated in the example in Fig. 2, the reconstructed Laue patterns are further processed using peak fitting and indexation software in order to obtain the lattice orientation for every ~1 $\mu m^3$ volume element along the path of the incident beam. The angular resolution for orientation mapping is of order ~0.01° for regions without large microstructural deformations at the micron length scale. After completing a depth-resolving DAXM wire scan at a particular sample position, the sample is translated ~1 μm perpendicular to the microbeam and another wire scan is obtained. Thus, by combining 2D scanning of the sample position with the DAXM wire scan, a full 3D orientation mapping with ~micron spatial resolution in all directions is obtained. Since this mapping produces independent, point-to-point measurements in all directions, high-resolution *intragrain* as well as *intergrain* orientation changes are directly determined.

## Results and Discussion

Aluminum slices were cut, lightly etched in a sodium hydroxide solution to remove cutting damage and mounted as indicated in Fig. 1 with the rolling direction (RD) along the surface normal. The beam was positioned near a sample edge with the incident beam typically passing through ~100 μm of aluminum. Beam attenuation with depth is not a major concern for locating diffracted peaks since the 1/e absorption distance in Al is approximately 1 mm for 20 keV x-rays. Initial measurements from as-rolled (200°C) Al samples revealed a typical grain size of ~5–10 μm and generally showed no evidence for streaking or broadening of diffraction peaks due to plastic deformation within individual grains. Thus the aluminum has recrystallized to the point where micron-sized volumes typically yield single-crystal Laue diffraction patterns. A preference for alignment with Al<001> along the RD was observed in typical Laue patterns, consistent with the presence of a cube texture component. Initial tests using the heating stage revealed little or no changes in grain morphologies for annealing 1 hour at temperatures below ~350ºC, and relatively rapid grain growth after annealing above 360ºC. When determining changes due to grain growth, it is essential that the same sample volume is reproducibly located before and after heating and cooling cycles. Since the sample position shifts during heating, small fiducial notches were cut in the edge of the samples using focused ion beam milling. These notches could easily be located with the x-ray beam and provided reproducible sample positioning within the ~1 μm spatial resolution.

Having established the temperature range where significant grain boundary motion was observed, the DAXM approach was used to obtain a point-by-point mapping of the orientation of each micron-sized volume element (voxel) within a randomly selected region in a hot-rolled aluminum sample. After low-temperature annealing at 250°C to remove hysteresis in the sample stage, a 10-μm × 10-μm × ~100-μm volume was scanned with 1 $\mu m^3$ voxels, requiring ~40 hours of measurement time. The scanned volume is approximately a rectangular prism. Scanning speed is currently limited entirely by CCD detector readout time (several seconds) and is not limited by the x-ray exposure time (~0.1 sec) for each image. Since area detectors with much faster electronics are now becoming available, the time required for a 3D volume scan should soon decrease by at least an order of magnitude. The sample was subsequently heated for 1 hour to 350°C, cooled to room temperature, and another 3D scan of the same sample volume was obtained. This procedure was repeated after annealing at temperatures of 355º, 360º, and 365ºC. After the higher-temperature anneals, grains were clearly much larger and additional scans covering larger volumes and using larger steps were also obtained. The raw microdiffraction images were reconstructed and indexed using the automated software described above. For every micron-sized voxel, the analysis produces a 3 × 3 matrix describing the lattice orientation. These orientation matrices provide the local information needed to construct detailed maps of the grain sizes, shapes and misorientations within the polycrystalline sample. During the data analysis, distinctive reconstructed Laue images containing rows of a large number of sharp Bragg peaks were observed at a few isolated locations superimposed on the fcc Al patterns. These anomalous patterns most likely indicate the presence of small (~1 μm), brittle second phase Al-Fe-Si precipitates known to be present in this sample [17]. Observation of these precipitates demonstrates the sensitivity of the DAXM technique to second phases and their effect on grain evolution will be examined in detail as results are analyzed [18,19].

Fig. 3 shows orientation maps of the same 2D slice taken from the center of the scanned 3D volumes after the 250°C anneal (Fig. 3a) and after three of the higher-temperature annealing steps (Fig. 3b,c,d). In each map, different shades for each pixel correspond to different Al[001] orientations. A thick line between pixels indicates a misorientation of greater than 20° and a thin line indicates a misorientation of between 5° and 20°. The initial hot-rolled microstructure consists

of many grains ~5–10 μm in size, and a large number of low-angle boundaries are observed in this (001)-textured sample. As the sample is annealed, both low-angle and high-angle boundaries are observed to change. After annealing at 350ºC, small changes can be noted, but most of the grains remain intact. After annealing at 355ºC, many small grains have been consumed, and only a few of the original grains are unchanged. After annealing at 360ºC, the scanned volume contains only three large grains meeting at a triple junction with relative misorientations greater than 20°. It is important to remember that the maps in Fig. 3 show only one of the 2D slices within the scanned 3D volume. Detailed data analysis including full 3D correlations is currently in progress.

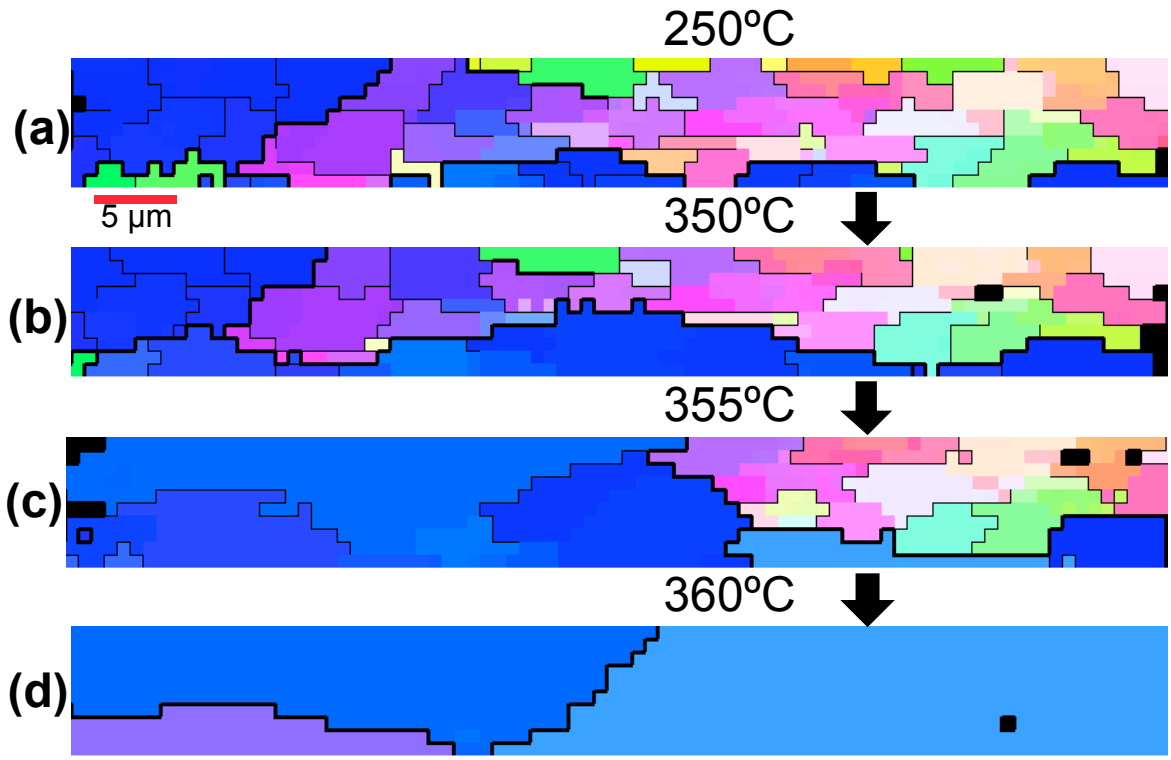


**Fig. 3** Grain evolution in polycrystalline aluminum. Orientation maps: after anneal at (a) 250ºC, and after annealing 1 hr at (b) 350ºC, (c) 355ºC, and (d) 360ºC. Each strip is a 2D slice taken from within a 3D x-ray microdiffraction scan. Bold lines indicate boundaries with misorientation greater than 20º, and thin lines show boundaries between 5º and 20º.

## Summary

Using newly developed 3D, nondestructive, microdiffraction techniques, an experimental "3D movie" consisting of frames showing grain evolution within polycrystalline aluminum during thermal annealing can now be obtained with micron spatial resolution. These measurments will provide detailed information describing the movement of *each* grain boundary and how it depends on the local environment – and thus provide much stronger tests of grain-growth models than averaged values of grain sizes and shapes. Detailed data analysis is currently in progress and future developments will include new techniques, faster data acquistion and new software. However, it is clear from these initial results that DAXM measurements will provide detailed 3D information useful for testing theories and computer modeling of grain growth in bulk materials.

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