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3-Dimensional Characterization of Polycrystalline Bulk Materials Using High-Energy Synchrotron Radiation

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Abstract. The implementation of 3-Dimensional X-Ray Diffraction (3DXRD) Microscopy at the Advanced Photon Source is described. The technique enables the non-destructive structural characterization of polycrystalline bulk materials and is therefore suitable for *in situ* studies during thermo-mechanical processing. High energy synchrotron radiation and area detectors are employed. First, a forward modeling approach for the reconstruction of grain boundaries from high resolution diffraction images is described. Second, a high resolution reciprocal space mapping technique of individual grains is presented.

Introduction

The microstructure of polycrystalline materials is characterized by a hierarchical arrangement of crystalline elements (grains, microbands, and subgrains). The arrangement is often highly heterogeneous, especially with respect to the dynamics during processing. Conventional experimental methods either lack sufficient spatial resolution or are surface or thin foil probes, which require samples to be sectioned before investigation to obtain spatially resolved results representative of bulk behavior. This destructive procedure prohibits studies of the dynamics of the individual elements. Thus, there is a need for a nondestructive method that provides comprehensive structural information for each of the crystalline elements within macroscopic volumes of the material. Furthermore, the method should be sufficiently fast to record the dynamics of 10–1000 elements simultaneously during processing.

Three-dimensional x-ray diffraction (3DXRD) microscopy is an emerging method that aims to fulfill these requirements. [1,2] The method is distinguished by two principles. The first is the use of a beam of high-energy x-rays generated by a synchrotron source for transmission studies. Hard x-rays (in the range 50–100 keV) can penetrate 4 cm of aluminum or 5 mm of steel. The second principle is a "tomographic" approach to diffraction. The conventional approach for providing spatially resolved information with diffraction is to scan the sample with respect to the beam. However, probing the sample point-by-point is generally too slow for dynamic studies. Hence, it has been replaced by an approach that provides information on many parts of the material simultaneously.

3DXRD microscopy has been developed at the European Synchrotron Radiation Facility (ESRF, France) by a group from the Risø National Laboratory (Denmark). Here we report on activities establishing 3DXRD capabilities at the Advanced Photon Source (Argonne National Laboratory)

within the high-energy program at beamline 1-ID. [3] Two case studies are presented. First, a forward modeling approach is being developed to extract the orientation and boundary topology of individual grains from high resolution, real space diffraction images. Second, a high resolution reciprocal space mapping technique is described. Both approaches are complementary to work performed at the ESRF.

Approach 1: Grain boundary mapping

The grain boundary network in polycrystalline materials is a critical determinant of materials properties. [4] The five parameter, mesoscopic specification of grain boundary character (three parameters describing crystallographic misorientation and two specifying boundary orientation) has recently been shown to yield an appealing interpretation of steady state grain boundary character distributions in several materials systems. [5,6] During grain growth, the populations tend to become inversely related to the associated grain boundary energies. Three dimensional computer simulations [7,8] yield populations related to energy through the Boltzmann factor. Because existing experimental methods for measuring grain boundary character are surface sensitive, there are no data addressing how these distributions arise on a grain by grain basis. Here, progress towards non-destructive measurements is described that will allow direct measurements inside of bulk samples. These measurements can probe dynamics, either in stop-action or in real time. The methods can also be applied to many other materials problems including non-destructive measurements during phase transformations and in composite materials.

Fig. 1 shows a schematic of the experimental apparatus. [9] A line focused (about 1.5 μm high by 1.3 mm wide) high energy (50keV) x-ray beam is used to illuminate a planar section of the sample. CCD images of diffracted beams are collected as the sample rotates through a one degree integration interval. Adjacent intervals are measured so as to collect as many Bragg peaks as possible from each grain in the illuminated plane. Data are collected over two 45 degree "wedges" in ω whose centers are 90 degrees apart. The shapes of diffraction spots are, ideally, projections along the diffracted beam of the shapes of diffracting grains. Because the Bragg angles are small, these projections are quite anisotropic yielding better reconstructed shape dimensions in the y-direction than x. Isotropic resolution is recovered through the use of two wedges.

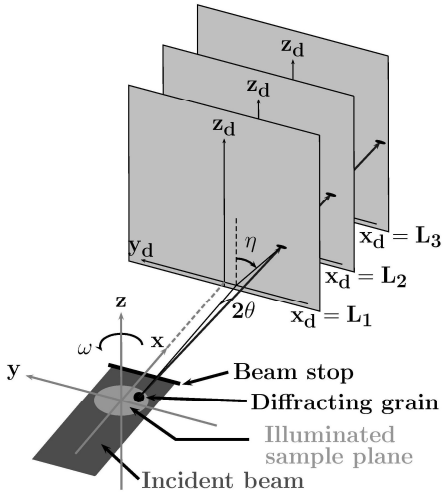

Figure 1. Schematic diagram of the grain mapping measurement. The sample rotates by ω about the z-axis which is perpendicular to the incident beam plane. High resolution (4 μm pixels) CCD images are collected at each of three rotation axis-to-detector distances, L, so that beams can be tracked through space. 2θ and η specify the direction of diffracted beams.

A forward modeling approach has been developed to reconstructing microstructure.[10] Raw CCD images are treated for noise reduction and diffraction spot identification. Each spot is thresholded at a fraction of its peak height to generate a binary data set. A simulation program then attempts to

define the microstructure. The sample plane is covered by a triangular grid. The crystallographic orientation of each area element is adjusted and optimized by a Monte Carlo process in an attempt to maximize overlap of its Bragg diffraction with experimentally observed intensity. An iterative procedure refines the area elements and focuses on boundary regions where the orientation changes rapidly. This approach has significant advantages over data inversion algorithms: i) for each area element, all relevant Bragg peaks at all measured ω are used in the optimization. ii) Monte Carlo optimization of experimental parameters can be done concurrently, iii) instrumental effects such as finite beam bandwidth and focusing geometry can be included, and iv) models of scattering from defected materials can be incorporated. In a verification exercise, it has been shown that a well-defined sub-area of a silicon single crystal can be reconstructed to about 10 μm resolution.[10]

Three of the first successful reconstructions of a polycrystal microstructure are shown in Fig. 2. These images show that independent data sets and independent fits yield consistent orientation locations and reasonable grain shape variations with z. However, the CCD image data used here is quite noisy and does not include higher order diffraction peaks that can better pin down grain shapes. Hence, some regions inside the sample do not satisfy the fit convergence criteria and the details of grain shapes (and, therefore, grain boundary character) cannot be reliably determined. With improved counting statistics and other hardware upgrades currently underway, reconstruction reliability is expected to improve substantially. A furnace has been constructed that is compatible with the diffraction geometry and will enable *in situ* annealing experiments.

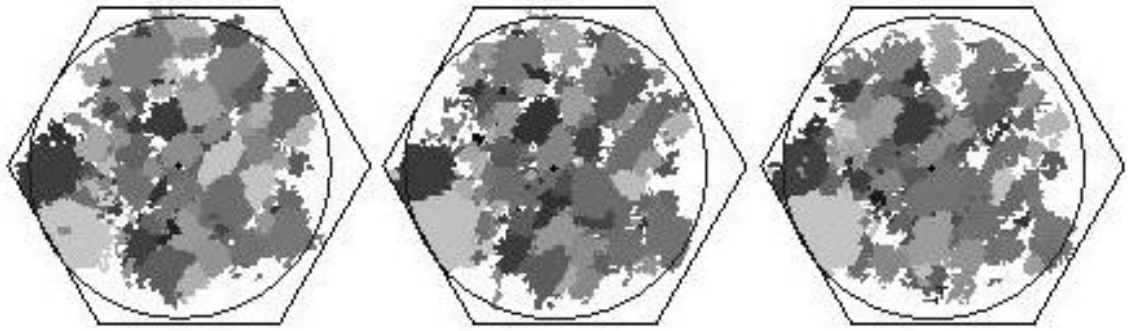

Figure 2. Three successive layers, obtained by translating the sample along z, of an annealed Al-1050 alloy reconstructed by forward modeling. The layers are 20 μm apart. The hexagons (600 μm side lengths) show the simulated region of sample space while the circles indicate the 1mm nominal diameter of the sample. The shaded regions show grains with the shading being determined by the local crystallographic orientation. Regions with no converged orientation are white. Reconstructed areas outside of the nominal sample diameter are within the estimated shape error.

Approach 2: High resolution reciprocal space mapping

Dislocation formation plays an important role in the plastic deformation of metals. The involved lengths scales are typically on the order of 1 μm and below. This suggests observation in reciprocal space as such features cannot be resolved by the present real space resolution of 3DXRD. Furthermore, the diffraction contrast of dislocations arises from their strain fields and results in characteristic broadening of reciprocal space intensity distributions (see e.g. [11]). Conventional line profile analysis is used to separate size and strain effects. However, line profiles arise as averages of diffraction from grains of different orientation and possibly different strain state and microstructure. In contrast, the technique presented here enables the measurement of the 3-dimensional reciprocal space intensity distributions from individual bulk grains and therefore provides a much more direct test of deformation structure models.

Approach 2: Experimental setup and procedure. At the 1-ID beamline of the APS specialized optics provide a vertically focused beam down to a size of 15 μm. [12] The typical working energy is 52 keV and the narrow energy bandwidth of $\Delta E/E = 10^{-4}$ enables a reciprocal space resolution down to 0.0005 $Å^{-1}$, which is an order of magnitude better than that attainable by electron microscopy. This is achieved by recording the diffraction patterns on an area detector which is positioned about 4 m behind the sample. The experimental setup is sketched in Fig. 3a. The samples are mounted in a tension rig that is attached to a xyz translation stage which in turn is mounted on a 3-axis diffractometer. Before high resolution profiles are recorded the orientations of the investigated grains and their distance from the sample surfaces is determined. Therefore a second area detector is temporarily inserted close behind the sample so that several complete diffraction rings are recorded. A set of rotation images is recorded by stepwise rotation around the phi axis. The orientation matrices of the diffracting grains are extracted by the multigrain indexing program GRAINDEX. [9]

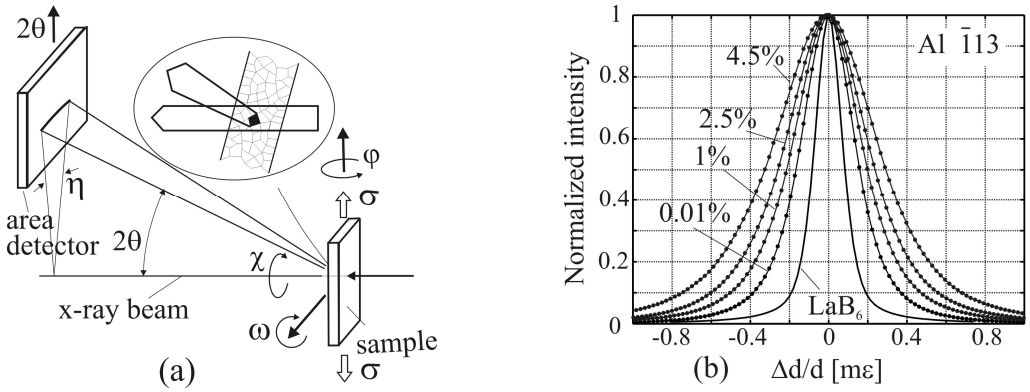

Figure 3. (a) Sketch of the experimental setup. The orientation angles φ, ω and χ are defined. The inset sketches the diffraction from an internal grain. σ indicates the direction of the applied tensile stress. Only the high resolution detector is shown. (b) Radial peak profiles from a single embedded Al grain, plotted in strain units. Also plotted is the profile of a LaB_6 calibration powder.

Experiments were performed on Al (AA 1050) and pure Cu specimens of several 100 μm thickness and at least ten average grain diameters across the thickness.

Approach 2: Results and discussion. In Fig. 3b axial peak profiles from an individual bulk grain are shown for tensile loads up to 4.5 %. The increasing broadening is obvious and a slight asymmetric tail towards smaller d-spacings is discernible. The asymmetric broadening of radial peak profiles from individual grains is understandable in terms of internal stresses arising in deformation structures: dislocation boundaries act as harder obstacles and develop forward internal stresses. [13] The asymmetry of several measured 113 reflections is found to depend not only on the (polar) angle between load axis and reciprocal lattice vector but also on the azimuth angle around the load axis. [14] This is not compatible with the asumption of an transversely isotropic subgrain structure which develops in [1 0 0] oriented grains. [13] It is explained [14] by the existence of single sets of parallel, extended dislocation boundaries which are aligned with a {111} plane and have been observed by TEM. [15]

By integration of the 3D reciprocal space intensity distribution along the radial direction orientation distribution projections are obtained. The 3-dimensional orientation distribution function of an individual deformed grain has been reconstructed from such projections. [16]

In Fig. 4 a series of raw images are plotted for small rotation intervals around the ω-axis. Distinct sharp spots (one is exemplary framed by a square) are discerned on a smooth background.

The spots are identified as originating from individual subgrains, whereas the background cloud of intensity is tentatively identified as arising from the disordered dislocation boundary regions. Measurements have been made under stepwise loading as well as under continuous loading. The direct observation of subgrains enables the determination of intra and inter-subgrain strains. The subgrain interiors are found to be strain free within the experimental accuracy but significant inter-subgrain strains are observed. These findings are in contradiction to some assumptions of the above mentioned 'composite model'. [13] Furthermore, the dynamics of individual subgrains have been observed during deformation. [12]

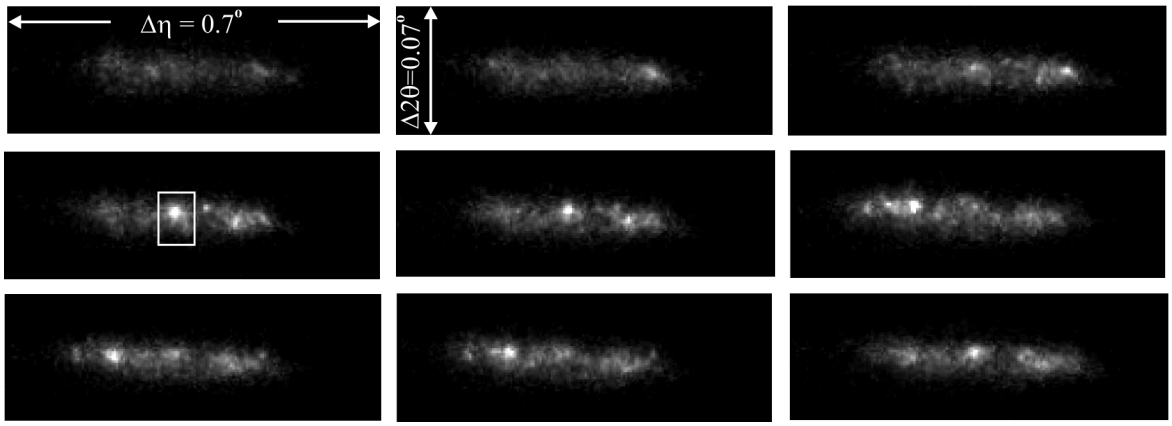

Figure 4: Series of nine raw diffraction images each representing a 2D reciprocal space profile of a 400 reflection from one grain at ~2% deformation. Each image is an integration over $\Delta\omega = 0.007$ deg. By stacking up these nine layers, the full 3D reciprocal space profile can be viewed. The angular widths of the marked spot were $\Delta 2\theta = 0.005$ deg and $\Delta\eta = 0.02$ deg, equivalent to a width of about 0.002 $Å^{-1}$ in all reciprocal space directions.

Outlook

Recently the 1-ID beamline at the APS has been dedicated exclusively to high energy x-ray diffraction. This provides the opportunity for dedicated instruments. In a first step planed for summer 2006 a dedicated setup for 3DXRD grain boundary mapping will be installed. Substantial improvements of efficiency and data quality are expected. In a second step the current end station will be rebuilt enabling full exploitation of the ample potential the APS offers for high energy diffraction. Further progress is expected from advances in area-detector technology and data acquisition and evaluation software.

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