2D and 3D X-Ray Structural Microscopy Using Submicron-Resolution Laue Microdiffraction

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ABSTRACT

We have developed a scanning, polychromatic x-ray microscopy technique with submicron spatial resolution at the Advanced Photon Source. In this technique, white undulator radiation is focused to submicron diameter using elliptical mirrors. Laue diffraction patterns scattered from the sample are collected with an area detector and then analyzed to obtain the local crystal structure, lattice orientation, and strain tensor. These new microdiffraction capabilities have enabled both 2D and 3D structural studies of materials on mesoscopic length-scales of tenths-to-hundreds of microns. For thin samples such as deposited films, 2D structural maps are obtained by step-scanning the area of interest. For example, 2D x-ray microscopy has been applied in studies of the epitaxial growth of oxide films. For bulk samples, a 3D differential-aperture x-ray microscopy technique has been developed that yields the full diffraction information from each submicron volume element. The capabilities of 3D x-ray microscopy are demonstrated here with measurements of grain orientations and grain boundary motion in polycrystalline aluminum during 3D thermal grain growth. X-ray microscopy provides the needed, direct link between the experimentally measured 3D microstructural evolution and the results of theory and modeling of materials processes on mesoscopic length scales.

INTRODUCTION

The availability of intense, highly-collimated x-ray beams at synchrotron facilities is enabling the ongoing development of a broad range of high-resolution x-ray structural microscopy techniques worldwide [1-3]. Various approaches are progressing rapidly, including x-ray techniques based on fluorescence, absorption, phase contrast or diffraction contrast. In general, hard x-rays (> 5 keV) are more penetrating than electron probes and hence can provide complementary nondestructive information from thicker samples or microstructures in the interior of bulk materials. In the approach described here, achromatic Kirkpatrick-Baez (KB) mirrors are used to focus white (polychromatic) radiation to submicron diameter, and x-ray Laue diffraction patterns are used to determine the local lattice structure, orientation and strain [4-8]. White-beam diffraction differs from more-conventional monochromatic scattering in several ways. First, many reflections comprising a full Laue diffraction pattern at a particular spatial position are collected simultaneously by an area detector in one image rather than during diffractometer step-scans. Second, no sample rotations are required to obtain diffraction information, eliminating “sphere of confusion” errors inherent when rotating individual grains in a polycrystalline material [4]. Instead, our approach is a scanning technique and requires only sample translations to obtain spatial resolution. As will be described below, measurements can

cover 2D areas (e.g. films) or 3D volumes (e.g. polycrystals). The x-ray microscopy technique we have developed is particularly well-suited for investigating microstructural features with length scales of tenths to hundreds of microns, that is, mesoscopic structures. On this scale, most materials are polycrystalline, with heterogeneous defects such as grain boundaries or second-phase precipitates playing essential roles in determining the mechanical, electronic, optical and magnetic properties. Multiscale computer simulations are increasingly being used to study the evolution of these heterogeneous structures, and experimental input is needed. Detailed x-ray microscopy measurements will provide valuable tests for computer models and will aid in developing our basic understanding of classic materials processes such as grain growth, fracture and plastic deformation.

EXPERIMENT

The microdiffraction experimental setup developed at the UNICAT beamline (sector 34) at the Advanced Photon Source is shown schematically in figure 1. White radiation (~8-20 keV) from an undulator is focused by glancing-angle reflection from a crossed pair of elliptically-figured KB mirrors. The mirrors nondispersively focus the beam to a submicron diameter (~0.5-1μm FWHM) near the sample position. As the beam penetrates a polycrystal sample, each grain acts as a single crystal and scatters a set of diffracted beams with energies satisfying the Bragg condition. The resulting Laue diffraction patterns are measured using a charge coupled device (CCD) area detector located at 90° to the incident beam.

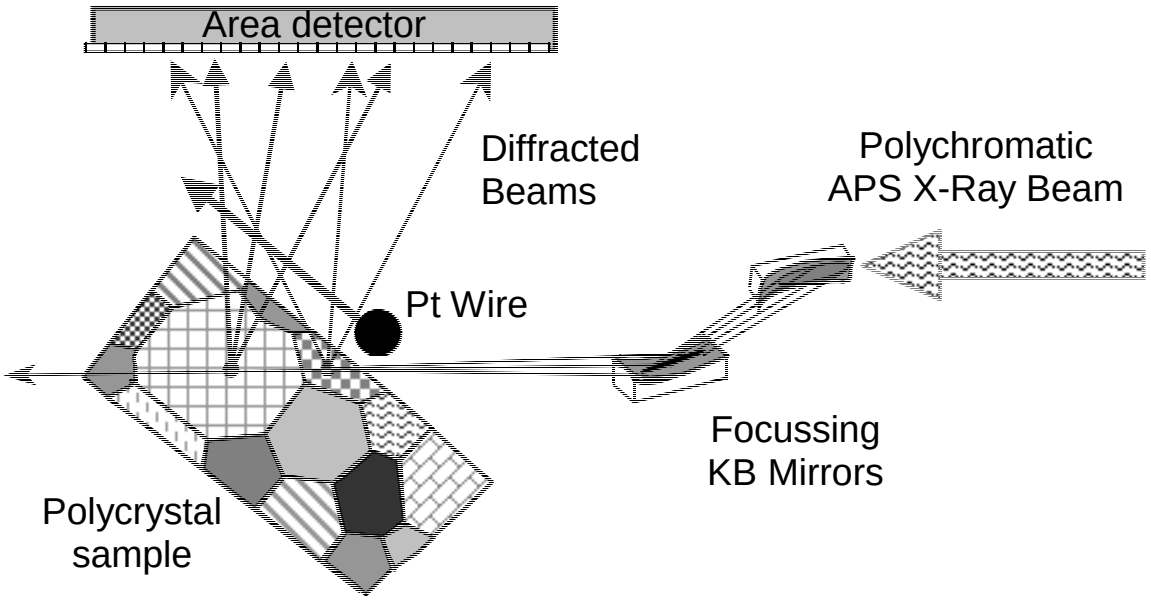

Figure 1. Schematic illustrating components of x-ray structural microscopy setup on the UNICAT beamline. Polychromatic synchrotron radiation is focused by a pair of KB mirrors onto a polycrystalline sample and diffracted beams are measured by the area detector.

If the sample is **2D**, such as a thin film, then generally only one grain will be in the path of the incident beam, and a simple single-crystal Laue diffraction pattern with peaks corresponding

to reflections from lattice planes will be generated. Automated computer analysis then fits peaks, indexes the pattern, and calculates the local crystallographic orientation and strain information at the particular location in the 2D sample [9]. By translating the sample and measuring the diffraction pattern at each spatial position, orientation and strain maps revealing the 2D grain microstructure can be obtained [7]. Conceptually, 2D orientation maps generated by step-scanned x-ray microdiffraction are very similar to maps generated by electron backscatter diffraction (EBSD) techniques [10]. Advantages of using x-rays rather than electrons include high angular resolution (~0.01°), the ability to make measurements in air, and the lack of charging effects with insulating materials, while the principal disadvantages are the relatively limited availability and high cost of a synchrotron beamline.

If the sample is a bulk **3D** polycrystal as shown in figure 1, the x-ray beam can penetrate a large number of grains, and the Laue patterns from all grains will be superimposed in the detector. In such cases, depth resolution is accomplished using a differential aperture x-ray microscopy technique [6]. The Pt wire acts as an absorbing knife-edge as a series of images are taken while the wire is translated in small steps parallel to the sample surface. By subtracting images taken at different wire positions, the Laue diffraction pattern corresponding to each particular depth along the incident beam can be uniquely reconstructed. The Laue patterns are then analyzed as in the 2D case to obtain orientation and strain information. Thus, measuring in a 3D step-scan array (2D translation of sample + 1D wire scan for depth) yields complete 3D orientation and strain maps for a particular sample volume. Since every ~1 μm^3 volume element is measured independently, intra-grain as well as inter-grain information is available. Conceptually, 3D x-ray microscopy maps are similar to maps generated by serial-sectioning EBSD [10]. However, the x-ray technique is nondestructive, and thus the microstructural evolution during materials processing (e.g. annealing) can be studied.

RESULTS

Several mesoscale materials investigations using the 2D and 3D x-ray microscopy techniques described above have been initiated at the UNICAT beamline [11,12]. Here, we demonstrate x-ray microdiffraction capabilities by presenting results from a 2D study of the epitaxial growth of oxide films and a 3D study of thermal grain growth in aluminum.

2D X-ray microscopy of epitaxial oxide films

Since high-angle grain boundaries suppress current densities in superconducting YBaCuO by orders of magnitude, significant research effort has focused on producing long lengths of highly textured YBaCuO coatings. In one approach, oxide buffer layers (e.g. CeO_2 and/or YSZ) and then superconducting films are grown epitaxially on recrystallized Ni foils which have a high degree of biaxial [001]<100> cube texture [13]. To help understand texture development in these materials, we have investigated the epitaxial growth of oxide buffer layers on roll-textured Ni foils during pulsed laser deposition (PLD) using x-ray microscopy [7]. In this case, two Laue patterns (substrate and film) are simultaneously measured at each spatial position. Figure 2 shows x-ray microbeam orientation maps from both the textured Ni substrate (Figure 2a) and a PLD-deposited CeO_2/YSZ buffer layer (Figure 2b) taken over a 0.72 mm by 0.72 mm area with 8 μm step size. Lighter-colored grains have their [001] axis closer to the surface normal. The grain structure in the oxide film is similar to that in the substrate, indicating approximately

epitaxial orientations. The angular information is shown more directly in pole-figure representations of the same data in Figures 2b (substrate) and 2e (film). Here, each pixel from the orientation map is used to generate a discrete point on the stereographic projection. Spatial information is missing, but the pole figures show the sharp cube texture and a 45° rotation of the in-plane film axes. Figures 2c (substrate) and 2f (film) are enlarged pole figures, zooming in to a radius of 5.3° around the surface normal. The zoomed figures reveal two important features. First, the texture of the film sharpens slightly due to tilts away from exact epitaxy and towards the surface normal. Second, the angular mosaic spread within individual grains is larger for film grains than for substrate grains, indicating local epitaxial disorder. The ability to simultaneously map the substrate and film orientation using x-ray microscopy yields important insight into the growth mechanisms and the resulting microstructures in these complex epitaxial systems. For example, microdiffraction results such as these can be used to develop atomistic models for understanding the deviations from exact epitaxy and for calculating percolation properties for superconducting films [7].

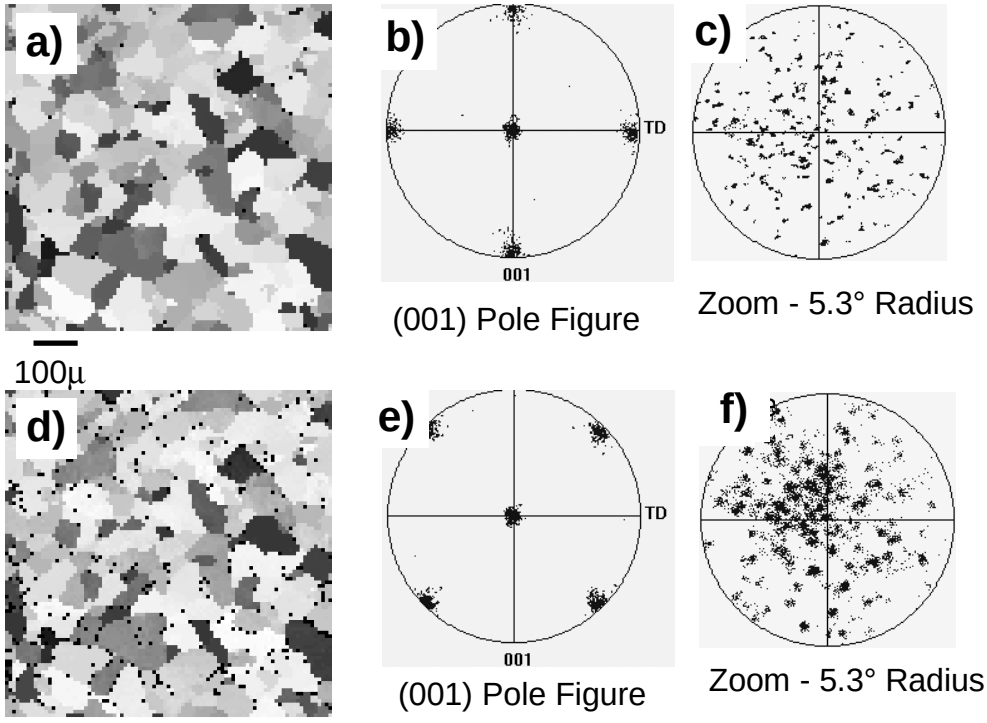

Figure 2 Top figures show a) an orientation map, b) the full (001) pole figure, and c) an enlarged central portion of the (001) pole figure from the Ni substrate. Bottom figures d), e) and f) show the same results from the oxide buffer layer film.

3D X-ray microscopy of thermal grain growth in Al

Grain growth during thermomechanical processing plays an important role in determining the physical properties of a wide range of materials. Grain sizes, shapes and orientations are intentionally controlled in many applications, ranging from fine-grained, high-strength steels to single-crystal, superalloy turbine blades. Large-scale computer models are emerging as valuable tools for understanding and predicting grain evolution during processing [14-16]. Experimentally, x-ray microscopy can now provide the first non-destructive 3D structural

measurements of grain growth in bulk polycrystals [17,18].

We have initiated in-situ x-ray microscopy studies of 3D grain growth in polycrystalline 1xxx series aluminum (~1% Fe, Si) obtained from Alcoa. The differential aperture x-ray microscopy technique was used to make a point-by-point 3D mapping of the orientation of each micron-sized volume element (voxel) within a well-defined (10- × 10- × ~100-μm) region in the as-received hot-rolled (200°C) aluminum. As illustrated in figure 3a, the initial grain size was approximately 5–10 μm. Figure 3b shows an expanded view where the 2D slices are separated. The sample was heated to induce grain growth, cooled to room temperature, and then re-mapped to measure the thermal migration of all grain boundaries. The technical issue of reproducibly locating the same sample volume after cooling was solved by ion milling small fiducial notches in the sample edge.

During initial observations, only small changes in grain morphologies were observed while heating below ~350ºC, and rapid grain growth was seen above 360ºC. Consequently, we obtained systematic measurements of the microstructural evolution after annealing (1 hr) at successive temperatures in the range of 350º - 365ºC. The data represents a "3D movie" consisting of frames showing the grain growth during thermal annealing. Detailed data analysis is currently in progress, but figure 3c shows one of the 2D slices at the beginning and at the end of the annealing sequence. The grains have clearly grown and are ~50 μm in size after annealing. During the annealing process, both low-angle and high-angle boundaries were observed to move. Most importantly, these results demonstrate that detailed experimental 3D grain growth data can now be obtained from bulk samples. When the data analysis is complete, experimental details will be compared quantitatively with computer models in order to test theories of 3D grain growth in polycrystalline materials.

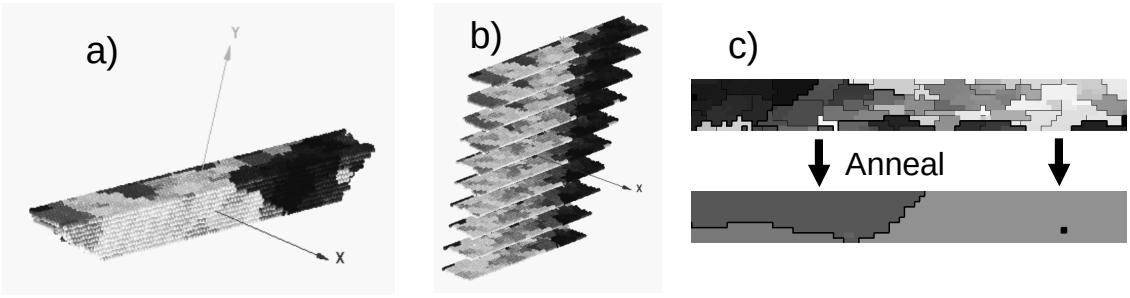

Figure 3 a) Initial microstructure of hot-rolled aluminum polycrystal. Each voxel represents 1 μm^3. b) expanded view of 2D slices. c) Microstructure evolution in a single 2D slice before and after the thermal annealing process.

CONCLUSIONS

Advances in synchrotron sources and x-ray optics have enabled recent progress in high-resolution x-ray microscopy techniques. Here, we have demonstrated structural microscopy using step-scan polychromatic Laue microdiffraction with high spatial and angular resolution. In 2D studies, x-ray microscopy yields orientation and strain maps, and thus provides valuable structural results for thin samples such as superconducting coatings, electronic devices, or thermal barrier coatings. In 3D studies, the differential-aperture x-ray microscopy technique

provides the first nondestructive 3D structural measurements in bulk materials with submicron point-to-point spatial resolution. This capability has been demonstrated in studies of 3D grain growth and will be applicable to many other mesoscale materials investigations.

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REFERENCES

1. B.C. Larson and B. Lengeler, editors, *Mat. Res. Soc. Bull.* **29**, 152 (2004).
2. X. Fu, H.F. Poulsen, S. Schmidt, S.F. Nielsen, E.M. Lauridsen, and D. Juul Jensen, *Scr. Mat.* **49**, 1093 (2003).
3. C. Riekel, *Rep. Prog. Phys.* **63**, 233 (2000).
4. G.E. Ice and B.C. Larson, *Adv. Eng. Mater.* **2**, 643 (2000).
5. N. Tamura, R.S. Celestre, A.A. MacDowell, H.A. Padmore, R. Spolenak, B.C. Valek, N. Meier Chang, A. Manceau, and J.R. Patel, *Rev. Sci. Instrum.* **73**, 1369 (2002).
6. B.C. Larson, W. Yang, G.E. Ice, J.D. Budai and J.Z. Tischler, *Nature* **415**, 887 (2002).
7. J.D. Budai, W. Yang, N. Tamura, J.S. Chung, J.Z. Tischler, B.C. Larson, G.E. Ice, C. Park and D.P. Norton, *Nature Mat.* **2**, 487 (2003).
8. W. Yang, B.C. Larson, J.Z. Tischler, G.E. Ice, J.D. Budai, and W. Liu, *Micron* **35**, 431 (2004).
9. J.-S. Chung and G.E. Ice, *J. Appl. Phys.* **86**, 5249 (1999).
10. C.T. Wu, B.L. Adams, C.L. Bauer, D. Casasent, A. Morawiec, S. Ozdemir and A. Talukder, *Ultramicroscopy* **93,** 99 (2002).
11. R.I. Barabash, G.E. Ice, N. Tamura, B.C. Valek, J.C. Bravman, R. Spolenak, and J.R. Patel, *J. Appl. Phys.* **93**, 5701 (2003).
12. W. Yang, B.C. Larson, G.M. Pharr, G.E. Ice, J.D. Budai, J.Z. Tischler and W. Liu, *Jour. Mater. Res.* **19**, 66 (2004).
13. D.P. Norton, A. Goyal, J.D. Budai, D. Christen, D. Kroeger, E.D. Specht, Q. He, B. Saffian, M. Paranthaman, C. Klabunde, D. Lee, B. Sales, and F. List, *Science* **274**, 755 (1996).
14. M.C. Demirel, A.P. Kuprat, D.C. George and A.D. Rollett, *Phys. Rev. Lett.* **90**, (2003), Art. 016106.
15. B. Radhakrishnan and T. Zacharia, *Modelling Simul. Mater. Sci. Eng.* **10**, 227 (2002).
16. K.G.F Janssens, *Modelling Simul. Mater. Sci. Eng.* **11**, 157 (2003).
17. J.D. Budai, W. Yang, B.C. Larson, J.Z. Tischler, W. Liu, H. Weiland, and G.E. Ice, *Materials Science Forum* **467-470,** 1373 (2004).
18. S. Schmidt, S. F. Nielsen, C. Gundlach, L. Margulies, X. Huang, and D. Juul Jensen, *Science* **305**, 229 (2004).