High Reciprocal Space Resolution 3-Dimensional X-ray Diffraction

B. Jakobsen[1], U. Lienert[2], J. Almer[2], H.F. Poulsen[1], W. Pantleon[1]

[1]Center for Fundamental Research: Metal Structures in Four Dimensions, Materials Research Department, Risø National Laboratory, Roskilde, Denmark;
[2]Argonne National Laboratory, Advanced Photon Source, Argonne, Illinois, USA;

Introduction

3-Dimensional X-Ray Diffraction (3DXRD) is an emerging tool for structural characterization in materials science (*e.g.* [1]). Based on the use of highly penetrating hard x-rays and a tomographic approach to diffraction, the method enables a non-destructive 3D description of the microstructural evolution within polycrystals. Under favorable conditions, hundreds of embedded grains can be characterized simultaneously with respect to position, shape, volume, crystallographic orientation and average strain tensor. With respect to studies of plastic deformation, the method has been used previously *e.g.* to characterize the rotation path of embedded grains as function of load [2].

An extension of the 3DXRD method towards high reciprocal space resolution is described here. The instrumentally challenging development is motivated by the expected decisive information on the evolving dislocation structure during deformation. From the atomic displacement field caused by the dislocation structure present in a crystal the diffracted intensity distribution in reciprocal space can be obtained via the kinematical scattering formalism. A large amount of work has been dedicated towards an interpretation of the reciprocal space intensity distribution in terms of a particular description of the dislocation structure (see *e.g.* [3]). Such an interpretation is complicated even further if the intrinsic reciprocal space intensity distribution is convoluted by a broad resolution function or averaged over orientation space.

The merits of the extension of 3DXRD to high reciprocal space resolution are revealed by comparison to conventional macroscopic peak profile investigations. Firstly, the measurements are specific to each grain. As such, the amount of averaging over various spatial heterogeneities is substantially reduced. The importance of such variations can be judged by comparing results from different grains. Likewise, the results obtained by conventional x-ray diffraction techniques are based on ensembles of grains with parallel reflecting lattice planes, but of different orientations around the lattice plane normals. Secondly, instead of measuring a few one-dimensional profiles, for each grain tens of 3D distributions are available, one for each reflection characterized. Hence, the information content in the data is substantially enhanced. This should allow for more reliable matching procedures to potentially more complex dislocation models, as well as for internal consistency checks, by comparing crystallographically equivalent entities.

Experimental setup

High energy synchrotron radiation is used to penetrate through bulk samples. The experiments were performed at the 1-ID-XOR beamline of the Advanced Photon Source (APS) at the Argonne National Laboratory. The setup is sketched in Fig. 1. A narrow bandwidth beam ($\Delta E/E<10^{-4}$) of typically 35 keV was provided by a flat Si double crystal monochromator. The main contributions to the instrumental resolution function arise from the energy bandwidth and from the experimental uncertainty of defining the angular spread of the diffracted beam. The latter is given by the grain size (or the size of coherently diffracting domains) and spatial resolution of the detector divided by the sample-to-detector distance.

Therefore a high reciprocal space resolution detector is placed at large distance from the sample, typically about 3.7 m. The scattering plane is vertical. As consequence of the large distance the covered azimuthal fraction of the diffraction ring becomes small, about $\Delta\eta = 7°$. Therefore a tension rig holding the sample is mounted on a 3-circle Eulerian cradle and reflections are centered on the high resolution detector by means of the rotations φ and χ. The third axis, ω, may be used to bring the tensile axis into a plane being perpendicular to the diffraction plane and containing the scattering vector.

In addition a second large area CCD is positioned closer to the sample. This detector samples all low-index diffraction spots arising from all illuminated grains. Data acquisition with this CCD serves as input to the multi-grain indexing program GRAINDEX [4]. Based on acquisitions during wide range φ-scans this program is able to sort the diffraction spots according to which grain they originate from and subsequently determine the orientation of the grains.

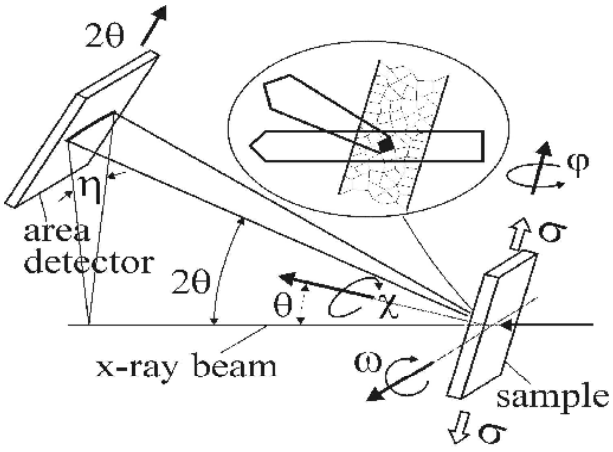

Fig. 1. Sketch of the experimental configuration with the stress-rig mounted on a Eulerian cradle. The orientation angles φ, χ and ω are defined. The insert sketches the diffraction from an internal grain. σ indicates the direction of the applied tensile stress. Only the high resolution detector is show.

Results

The developed setup is quite versatile, and work is presently in progress in several areas. The materials used for experiments are primarily commercial pure aluminum (AA1050) and 99.98% pure copper. The materials are recrystallized, giving the possibility to follow plastic deformation from its beginning. For each type of material appropriate dimensions of the tensile samples are chosen. The method has already produced interesting results on orientation and strain dependence of the

reciprocal space projections from one bulk aluminum grain after tensile loading [5]. In the following a second experiment performed on copper is reported [6].

The variance of the axial lattice strains of individual grains was measured during tensile loading of a weakly textured copper polycrystal. Peak profiles were measured for 440 reflections arising from 20 bulk grains, all of which were at least one average grain diameter below the surfaces within a 0.2 mm thick specimen. Raw images from a selected grain are shown in Fig. 2. The reflecting lattice planes were all perpendicular to the tensile axis. The variance in the measured axial lattice strain between grains of the same orientation can therefore be attributed to differences in the boundary conditions imposed by the different environments of neighboring grains. The results were compared to finite element modeling (FEM). Up to the highest applied strain of 2%, the experimental and predicted values were in fair agreement and the standard deviations may be roughly approximated as 6% of the average axial lattice strains (see Fig. 3).

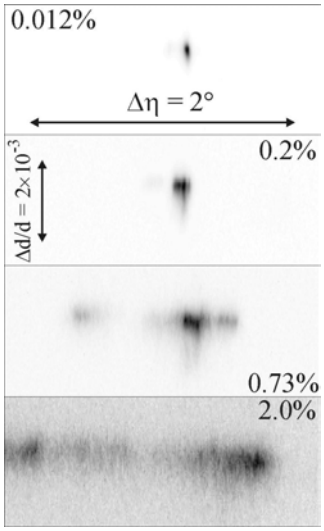

Fig. 2. Raw detector images of a selected diffraction spot. The tensile strains are indicated. The vertical axis—measuring the lattice strain—is enlarged by a factor of 5 against the horizontal direction, which is tangential to the diffraction ring. The diffraction spot to the left in the bottom image has rotated into the detector field during the last deformation step. The lattice strain and mosaicity scales are indicated.

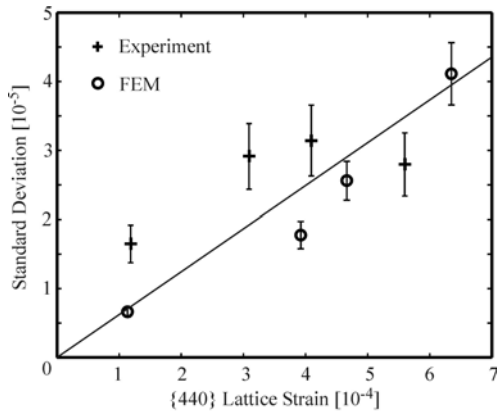

Fig. 3. Comparison of the experimental and FEM-simulated standard deviations of the {440} axial lattice strain components. The values are plotted versus the respective average axial lattice strains. A straight line obtained by linear regression is plotted as a guide to the eye.

Summary and Outlook

The extension of 3DXRD to high resolution in reciprocal space is a quite promising new technique. By analyzing the details of the diffraction spots information on processes internally in the grains can be gathered. The understanding of the development of subgrain structures during deformation is an important issue, and this technique could give the answer to many outstanding questions.

When metals are investigated after deformation it is observed that dislocations form substructures. A fundamental and unanswered question is if creation of these structures is part of the deformation dynamics, or is due to relaxation after the deformation has ended. Feasibility studies have shown that with the current setup it is possible to follow a single grain during deformation. By dynamically observing the peak shape of a single grain it will be possible to discriminate between the two possibilities. Such work is in progress.

The angular resolution in the directions perpendicular to the scattering vector is about 0.005°. With such a resolution it will be possible to observe individually diffracting subgrains. Establishing of such an ultra high resolution technique and their exploitation is therefore envisaged.

Acknowledgments

B. Jakobsen, W. Pantleon and H.F. Poulsen acknowledge the Danish National Research Foundation for supporting the Center for Fundamental Research, within which part of this work was performed, as well as the Danish Natural Science Research Council (SNF) for financial support via Dansync. Use of the Advanced Photon Source was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. W-31-109-ENG-38.

Part of the content of the report is extracted from a paper [6] that appears in a copyrighted publication.

References

[1] H.F. Poulsen, D. Juul Jensen, G.B.M. Vaughan, MRS Bull. **29**, 166-169 (2004).
[2] L. Margulies, G. Winther, H.F. Poulsen, Science **291**, 2392-2394 (2001).
[3] R.L. Snyder, J. Fiala, H.J. Bunge (Eds.), Defect and microstructure analysis by diffraction (Oxford University Press, Oxford, 1999).
[4] E.M. Lauridsen, S. Schmidt, R.M. Suter, H.F. Poulsen, J. Appl. Cryst. **34**, 744-750 (2001).
[5] W. Pantleon, H.F. Poulsen, J. Almer, U. Lienert, Mater. Sci. Engng. A **387-389,** 339-342 (2004).
[6] U. Lienert, T.-S. Han, J. Almer, P.R. Dawson, T. Leffers, L. Margulies, S.F. Nielsen, H.F. Poulsen, S. Schmidt, Acta mater. **52,** 4461-4467 (2004).