# The grain microstructure of polycrystalline materials as revealed by the combined use of synchrotron X-ray imaging and diffraction techniques

Wolfgang Ludwig, M. Herbig, A. King, P. Reischig, Henry Proudhon, Jean-Yves Buffiere

# The grain microstructure of polycrystalline materials as revealed by the combined use of synchrotron X-ray imaging and diffraction techniques

**W. Ludwig[a,b], M. Herbig[a], A. King[c], P. Reischig[d], H. Proudhon[e], J.Y. Buffière[a]**

[a] Université de Lyon, MATEIS, UMR5510, CNRS, France
[b] ESRF, BP 220, Grenoble, France
[a] GKSS, Geestacht, Germany
[d] Karlsruhe Institute of Technology, Germany
[5] Mines Paristech, Paris, France

## Abstract

Combining the principles of x-ray imaging and diffraction techniques, it has recently become possible to map the 3D grain microstructure in a range of polycrystalline materials. Associating this 3D orientation mapping with conventional attenuation and/or phase contrast tomography yields a non-destructive characterization technique, enabling time-lapse observation of dynamic processes in the bulk of structural materials. The capabilities and limitations., as well as future perspectives of this new characterization approach will be discussed and illustrated on selected application examples.

## Introduction

In the course of the last years significant progress has been made in the development of non-destructive, three-dimensional orientation imaging techniques combining the principles of X-ray imaging and diffraction techniques. In this paper focus will be given on a method termed X-ray 'diffraction contrast tomography' (DCT) (Johnson 2008, Ludwig 2009) , currently developed at the European Synchrotron Radiation Facility (ESRF). In contrast to established orientation imaging techniques like electron backscatter diffraction (EBSD) or (polychromatic) X-ray Laue micro diffraction, where spatial resolution is determined by the interaction volume of a point focused electron or X-ray beam, DCT follows a different strategy: the whole gage volume is illuminated by an extended, monochromatic X-ray beam and the transmitted as well as diffracted beams are recorded simultaneously on a high resolution detector system, which in turn determines the maximum achievable spatial resolution. The analysis of these compound images provides the 3D attenuation coefficient distribution, as well as the (average) crystallographic orientation, elastic strain tensor and the 3D shape of the grains in the illuminated sample volume. The outlined 3D acquisition mode leads to a tremendous gain in acquisition speeds since it involves only a one-dimensional scanning procedure around an axis perpendicular to the X-ray beam. This, in turn, offers interesting possibilities to perform time-lapse observations of grain microstructures subject to increments in plastic deformation.

## Experimental

DCT is a monochromatic beam rotation technique, combining the principles of 3D X-ray diffraction microscopy (Poulsen 2004) and X-ray micro tomography. During a 360 degree continuous rotation of the sample, each grain runs through a series of diffraction alignments, giving rise to diffracted beams. Part of these diffraction spots are captured on the high resolution imaging detector system, positioned closely behind the sample (Figure 1). Like in conventional micro tomography, one can determine the three-dimensional distribution of the X-ray attenuation coefficient from the attenuation in the transmitted beam. The analysis of the diffracted beams provides access to the crystalline microstructure. The processing route (Ludwig 2009) is summarized in Figure 2. After segmentation, Friedel pairs of diffraction spots (hkl and -h-k-l reflection from the same grain) are automatically identified ("pair matching"). A polycrystal indexing algorithm based on the analysis of such pairs of diffraction spots (Reischig 2008) classifies the diffraction spots according to their grain of origin and determines the average orientation and elastic strain state of the grains.

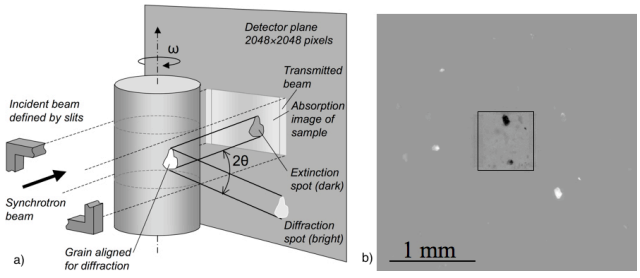


*Figure 1. left: DCT shares a common setup with X-ray microtomography. The transmitted as well as (part of the ) diffracted beams are captured on the high resolution detector positioned closely behind the sample.*

The 3D shape and the position of the grains in the sample volume are determined with the help of algebraic reconstruction techniques, using the 2D diffraction spots as parallel projections of the unknown 3D grain volume (Figure 2, bottom right).

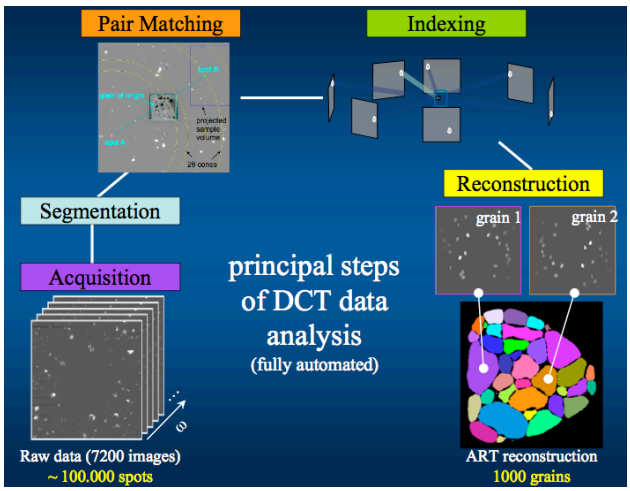


*Figure 2 Principal steps of the DCT analysis procedure*

## Application examples

In the course of the past years, the technique has been applied to a variety of case studies in different classes of materials. A non-exhaustive list of ongoing projects includes the analysis of:

- stress corrosion cracking in stainless steel (King 2008)
- plastic deformation in Al alloys
- creep deformation in snow
- grain growth in Al, Ti, and $SrTiO_3$
- elastic deformation in Ti alloys (Reischig 2008)
- short fatigue crack / microstructure interaction in Ti and Mg alloys (Herbig 2010, King 2010)

In order to illustrate the capabilities of the combined imaging and diffraction methodology we briefly discuss first results obtained from the latter two studies.

### 1) Grain resolved determination of type 2 elastic strains

The analysis of Friedel pairs of diffraction spots provides an angular accuracy in the determination of the scattering vector of order of $5 \cdot 10^{-4}$ rad. During a full rotation, typically several tens of independent reflections are acquired for each of the grains. Taking into account the magnitude and direction of the measured scattering vectors, one can assign an (average) elastic strain tensor to each of the grains. The result of this fitting procedure is shown in Figure 3a.

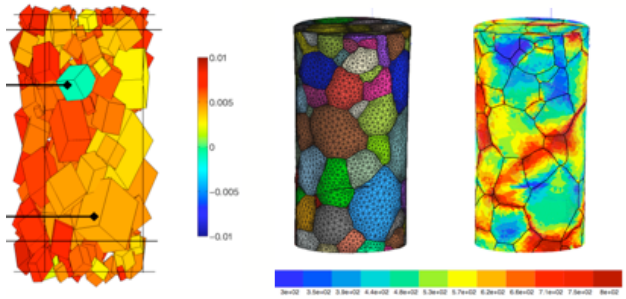


*Figure 3. a) Grain resolved representation of the elastic strain along of the tensile axis. b) Tetrahedral mesh representation of the measured microstructure c) Finite element prediction of the von Mises stress.*

Each of the grains is represented by a parallelepiped and colour coded according to the strain component in direction of the tensile loading axis. One can observe a left - right gradient, presumably caused by misalignment of the sample grips in the loading device.

After transformation of the 3D volume data into a finite element mesh (Figure 3b), it is possible to run numerical simulations of the deformation behaviour. Figure 3c shows the prediction of the van Mises stress obtained by finite element simulation based on the measured microstructure. The elastic anisotropy of the individual grains leads to incompatibility stresses close to the grain boundaries. This behaviour underlines the importance of characterization techniques providing access to the spatial distributions of elastic strain at the sub-grain resolution length scale.

### 2) Observation of short fatigue crack propagation

DCT shares a common experimental setup with high-resolution X-ray phase contrast tomography (Cloetens 1997) - a technique which can reveal faint variations in electron density and which can be used to characterize damage evolution such as growth of short fatigue cracks in structural alloys. Figure 4 shows an example for the combined use of both techniques: the initial grain microstructure has been mapped by DCT and the subsequent growth of a fatigue crack (shown in white) has been monitored using phase contrast tomography (interrupted in-situ test on the synchrotron instrument). The local crack propagation rate and the orientation of the crack facets with respect to the grain orientation have been analysed and can be interpreted in terms of crystallographic slip system activity.

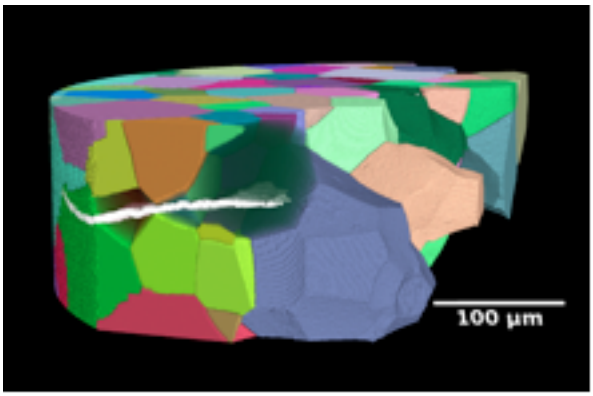


*Figure 4. A combination of X-ray phase contrast tomography (observation of crack propagation) and DCT (grain orientation and shapes) can be used for the observation of short fatigue crack propagation in the bulk of metallic alloys.*

## Limitations

The ultimate spatial resolution of the outlined DCT methodology is determined by the X-ray detector system and currently limited to values close to 1 µm. This in turn implies a minimum grain size of order of 10-20 µm. The sample diameter and beam size determine the total number of grains and have to be adjusted in order to limit overlap of diffraction spots on the detector system. In the case of solidification or recrystallization microstructures, typical sample diameters are about 20 times the average grain size

and up to 1000 grains and more can be analysed in these conditions. Note that in the presence of strong texture and/or orientation gradients the number of grains that can be analysed in a singel scan will reduce to 100 or less. DCT is currently available at the insertion device beamlines ID11 (Materials Science) and ID19 (Imaging) at the ESRF. Typical scanning times range between 30 min and 20h, depending on the spatial resolution and energy used.

## Perspectives

The presence of a grain substructure (orientation and strain gradients) results in local variation of the diffraction vector and lead to a variation of diffracted intensity as a function of rotation angle (Figure 5). At moderate levels of deformation simulations indicate the possibility to reconstruct these variations in the orientation and strain distributions by means of iterative reconstruction procedures. Switching from extended beam to line or pencil beam illumination modes, is a possible way to reduce the complexity of the reconstruction task and to extend the applicability range towards higher levels of deformation. Full field DCT characterization in the undeformed state may be regarded as a prerequisite for subsequent use of monochromatic beam scanning techniques: knowing the 3D shape, position and diffraction alignments of the grain(s) of interest the time consuming 2D or 3D scanning procedures can be restricted to the minimum required.

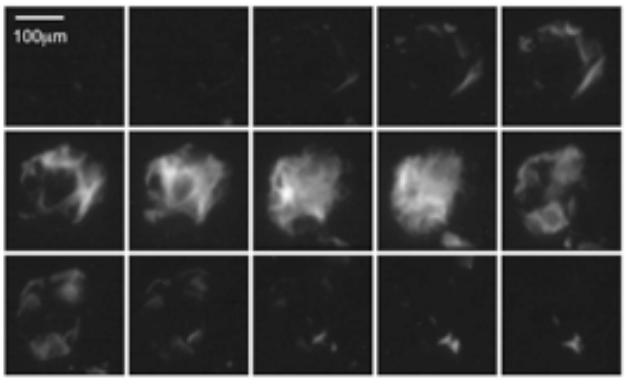


*Figure 5 Variation of diffracted intensity as a function of the rotation angle. The observed spread is a signature of orientation and strain gradients inside the grain.*

For higher values of plastic strain, a different data analysis strategies based on forward simulation may be more appropriate (Suter 2006). In the simplest case one would aim at the minimization of a cost function expressing the difference between the forward simulated diffraction pattern (based on the current approximation of the microstructure) and the experimentally observed diffraction patterns. Due to the high dimensionality (the full problem has 12 free parameters: orientation (3) elastic strain (6) and position (3)) such approaches are computationally very intense.

Last but not least it shall be noted that the current limitations in terms of spatial resolution may in some cases be extended to the sub-micrometer range by combining the concepts of X-ray microscopy and an earlier variant of diffraction contrast tomography, exploiting the extinction contrasts visible in the direct beam (Ludwig 2008).

## Conclusions

The combined x-ray imaging and diffraction methodology described in this paper provides access to a three-dimensional orientation imaging mode applicable to certain classes of polycrystalline materials. Sample volumes containing up to 1000 grains can be characterized in terms of local attenuation, 3D grain shape, crystallographic orientation and elastic strain tensors (average values per grain). Time lapse observations of the *evolution* of these variables in the bulk of the material during heat treatments and/or (limited) mechanical deformation have been demonstrated and offer the possibility to compare experimentally observed against numerically simulated material behaviour.

## References

H. F. Poulsen. *Three-dimensional X-ray diffraction microscopy*. Springer, 2004.

G. Johnson, A. King, M. G. Honnicke, J. Marrow, and W. Ludwig. *Journal of Applied Crystallography*, 41:310–318, 2008.

W. Ludwig, P. Reischig, A. King, M. Herbig, E. M. Lauridsen, G. Johnson, T. J. Marrow, and J. Y. Buffiere.. *Review of Scientific Instruments*, 80(3):033905–9, 2009.

P. Reischig. Determination of elastic strain tensors from x-ray diffraction contrast tomography. Master's thesis, Delft University of Technology, 2008.

A. King, G. Johnson, D. Engelberg, W. Ludwig, and J. Marrow. *Science*, 321(5887):382–385, 2008.

M. Herbig, A. King, P. Reischig, E. Lauridsen, J. Marrow, J. Buffiére, and W. Ludwig., in preparation.

W. Ludwig, S. Schmidt, E. M. Lauridsen, and H. F. Poulsen. *Journal of Applied Crystallography*, 41:302–309, 2008.

R. M. Suter, D. Hennessy, C. Xiao, and U. Lienert. *Review of Scientific Instruments*, 77(12):123905, 2006.