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X-ray diffraction microscopy: emerging imaging techniques for nondestructive analysis of crystalline materials from the millimetre down to the nanometre scale

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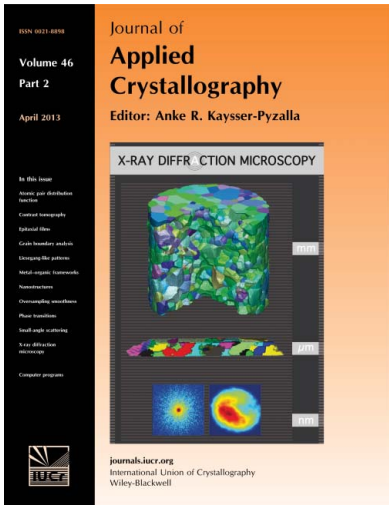
The discovery made in 1912 by Laue, Friedrich and Knipping, as well as the subsequent work by the Braggs, father and son, laid down the basis for one of the most successful experimental methods in the history of science: X-ray diffraction (XRD). The method has been essential to the scientific discoveries of more than 30 Nobel laureates, illustrating its utility and wide dissemination in many scientific disciplines, encompassing physics, chemistry, biology and medicine. In addition, XRD has underpinned many advances in applied sciences, for example, materials science in the fields of alloy development and component testing, as well as engineering sciences, thereby facilitating the contributions of both basic and applied sciences to the welfare of today's society. This has been highlighted by the recent decision of the United Nations to declare 2014 the International Year of Crystallography. Celebrating this achievement, a number of events and publications will review the major work carried out over the past 100 years; during this period XRD has evolved to become a powerful and indispensable tool in all those scientific fields where knowledge of the relations between crystal structure, function and properties plays an essential role. Today new sources and new techniques open new horizons in XRD techniques and science using XRD.

As part of the preparations for the International Year of Crystallography, *Journal of Applied Crystallography* has published a series of articles on novel diffraction methods enabling visualization of the internal structure of crystalline materials from the millimetre down to the nanometre scale. These works, now gathered together into a virtual issue on X-ray diffraction microscopy, summarize recent developments in the nondestructive monitoring of structural and dynamic information from the bulk of materials in three dimensions. It is believed that this coupled information could lead to a deeper understanding of some of the major problems in materials and engineering sciences, such as polycrystal plasticity or damage. The selected articles describe briefly the methods, focusing mainly on applications. Advantages, limits and future prospects of the techniques are also highlighted.



A synthesis of the methods developed around the concept of three-dimensional X-ray diffraction is presented in the article by Poulsen (2012): *An introduction to three-dimensional X-ray diffraction microscopy*. A common feature of these methods is the three-dimensional spatial character of the recovered information, usually attained through a tomographic approach. The main aim of the techniques is the complete spatial, dynamic and time-resolved characterization of a material. Resolution requirements, however, impose constraints on the diffraction setup or acquisition time, demanding that the experimentalist find a good compromise between the obtainable spatial, angular and time resolutions and those characteristic for the investigated phenomenon. Selection of a specific method is usually determined by the nature of the required information and the crystallographic perfection of the sample.

The granular structure of nearly perfect (undeformed) polycrystals can be reconstructed by three-dimensional orientation imaging microscopy (3D-OIM; Poulsen, 2012) or by diffraction contrast tomography (DCT; Reischig *et al.*, 2013). While 3D-OIM reconstructs the structure layer by layer, DCT uses a wide box beam ($\sim$1 $\times$ 1 mm) and builds up the polycrystal grain by grain, each grain being separately reconstructed. The DCT approach assumes that crystallographic orientation is constant through the grain,

which limits its applicability to undeformed crystals free of large orientation gradients. The presence of the latter has the effect of spreading the diffracted intensity over many detector frames, which can cause spot overlap. Intensity overlap is less probable in the case of a line beam ($\sim$1 mm $\times$ 1 μm), enabling the extension of the method to plastically deformed crystals. Subgrains formed during the grain subdivision process are shown by Poulsen (2012) and by Li *et al.* (2012), the latter implementing a forward modelling approach called near-field high-energy diffraction microscopy. The first *in situ* tests show encouraging evidence for the suitability of the methods at moderate degrees of deformation, but more work is needed in terms of validation and defining the limitations.

The increase of the spatial resolution of the reconstruction can be achieved in many ways: by utilizing magnifying optics, or by decreasing the pixel size of the detectors or the cross section of the beam. The latter solution was chosen in the case of polychromatic three-dimensional X-ray microscopy (P3DXM; Larson & Levine, 2013). The method uses a microbeam coupled with a differential-aperture (knife-edge) depth-profiling technique and enables the determination of local crystal orientations, grain morphologies and grain interface types, as well as the components of the deviatoric elastic strain tensor, with submicrometre spatial resolution in three dimensions. These unique features make P3DXM a powerful tool for fundamental investigations of deformation mechanisms through measurements of local elastic strain and geometrically necessary dislocation densities, which can be compared with predictions of existing theories.

Diffraction/scattering computed tomography (DSCT), presented by Álvarez-Murga *et al.* (2012), also uses a pencil beam which, however, is monochromatic. DSCT is well adapted for differentiating among multi-phase compounds with similar densities and compositions by exploiting their structural and/or microstructural properties. The term scattering additionally emphasizes the applicability of this technique for detecting poorly ordered phases. The principle of the method is based on the independence of the diffracted/scattered intensity from the beam direction, which implicitly limits its application to randomly oriented powder-like samples. The beam size and the corresponding spatial resolution should be therefore adapted to the grain size of the specimen, fulfilling the requirement that the number of crystallites in the irradiated volume is large. In spite of this constraint remarkable spatial resolutions of the order of 100 nm could be obtained for nanograined materials (Álvarez-Murga *et al.*, 2012). An interesting application is given by Stock & Almer (2012), by reconstructing the structure of an SiC-monofilament-reinforced Al-matrix composite.

The increase in spatial resolution of the reconstructed structures towards the nanometre scale is very attractive for materials science, nanoscience and biology. Coherent diffraction imaging (CDI) achieves this by exploiting the increased transverse coherency of the latest synchrotron sources and the power of ingenious iterative phase retrieval algorithms, which recover a unique real-space image from oversampled far-field diffraction intensities. A comparison of the most powerful algorithms applied to noisy diffraction patterns is presented by Rodriguez *et al.* (2013). They show that the newly proposed oversampling smoothness algorithm leads to increased consistency values when applied to the reconstruction of a weakly scattering biological cell. To overcome some limitations of CDI, an efficient ptychography method (by scanning an extended sample through a confined illumination and recording diffraction patterns at overlapped scanning positions) has been published recently. Huang *et al.* (2012) present a combined Bragg CDI with ptychography technique that allowed them to reconstruct the three-dimensional electron density and strain field of an extended ZnO microcrystal.

We appreciate the contributions of all authors to this virtual issue of *Journal of Applied Crystallography* as they have made an effort to reveal the basics, the current limitations and the great potential of X-ray diffraction microscopy. We expect that the present collection of articles will be of use not just to specialists but in particular to potential users, for example, in the selection of the most appropriate method for their specific scientific problem.

This special issue is available at http://journals.iucr.org/special_issues/2013/imaging/.

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