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## Mapping of individual dislocations with dark-field X-ray microscopy

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This article presents an X-ray microscopy approach for mapping deeply embedded dislocations in three dimensions using a monochromatic beam with a low divergence. Magnified images are acquired by inserting an X-ray objective lens in the diffracted beam. The strain fields close to the core of dislocations give rise to scattering at angles where weak beam conditions are obtained. Analytical expressions are derived for the image contrast. While the use of the objective implies an integration over two directions in reciprocal space, scanning an aperture in the back focal plane of the microscope allows a reciprocal-space resolution of  $\Delta Q/Q < 5 \times 10^{-5}$  in all directions, ultimately enabling highprecision mapping of lattice strain and tilt. The approach is demonstrated on three types of samples: a multi-scale study of a large diamond crystal in transmission, magnified section topography on a 140 µm-thick SrTiO<sub>3</sub> sample and a reflection study of misfit dislocations in a 120 nm-thick BiFeO<sub>3</sub> film epitaxially grown on a thick substrate. With optimal contrast, the half-widths at half-maximum of the dislocation lines are 200 nm.

#### 1. Introduction

Dislocations are typically studied by transmission electron microscopy (TEM). With atomic resolution, comprehensive information can be gathered on, for example, the strain field in a dislocation core (Dong & Zhao, 2010) or the 3D arrangement of dislocations in networks (Barnard *et al.*, 2006; Ramar *et al.*, 2010; Liu *et al.*, 2014). However, TEM is inherently limited to the study of thin foils. For non-destructive mapping of individual dislocations in the bulk, X-ray imaging is prevalent.

In conventional X-ray topography, a 2D detector or film is placed in the Bragg-diffracted beam downstream of the sample (Tanner, 1976). The diffracted intensity is projected onto a 2D image, a 'topograph'. This technique allows one to visualize long-range strain fields induced by the dislocations. 3D mapping can be provided in several ways. First results were achieved by preparing 'stereo pair' diffraction topographs (Lang, 1959; Haruta, 1965), which provide two views of the defects, and later approaches focused on recording a number of closely spaced 'section' topographs (Medrano et al., 1997; Ohler et al., 2000). Synchrotrons made more elaborate methods accessible. In topo-tomography, as presented by Ludwig et al. (2001), a large number of projections are obtained by rotating the sample about the scattering vector. By generalizing cone beam X-ray tomography, these can be reconstructed into a voxellated 3D model. Topo-tomography

has been used to map networks containing hundreds of dislocations. The spatial resolution, however, is inherently limited [see also Tanner (1976)], and was 10 µm in the study reported (Ludwig et al., 2001). In a similar manner, laminography has been successfully applied to studies of dislocations in wafers (Hänscke et al., 2012). The limitation on resolution was overcome in a study with a polychromatic nanobeam by Hofmann et al. (2013), where all six independent strain components were mapped around one single dislocation with a resolution of 500 nm. The drawback in this case is that the method involves scanning the nanobeam with respect to the sample, a procedure that is relatively slow; hence generalization to mapping an extended network in three dimensions is not trivial. Recently, studies of dislocations within isolated nano-sized crystals have also been done using X-ray coherent techniques (e.g. Ulvestad et al., 2017), but again generalization to bulk samples is not straightforward.

Here we demonstrate a new approach to the 3D characterization of defects within extended internal volumes of near-perfect single crystals, grains or domains. This is based on dark-field X-ray microscopy, where an X-ray objective lens is placed in the diffracted beam (Simons *et al.*, 2015; Simons, Haugen *et al.*, 2018), providing an inverted and magnified projection image on a detector in the imaging plane. The spatial resolution and field of view are a function of the magnification, which depends on the lens configuration and the sample-to-objective and objective-to-detector distances. Similarly to optical microscopy or TEM, the microscope is also associated with a Fourier/diffraction plane, the back focal plane. Detailed descriptions of the optical properties in the image plane and the back focal plane are given by Poulsen *et al.* (2017) and Poulsen *et al.* (2018), respectively.

In the following, we first summarize the acquisition geometry of dark-field microscopy. Next, we present two methods for mapping dislocations. One method is a magnified version of classical topography. In the other, an aperture is introduced in the back focal plane to define a (small) local region in reciprocal space. By scanning the aperture one can visualize the strain field around dislocations, for example with the aim of observing the interaction between these. We describe the optical principles and demonstrate the use of the methods by three examples. The first is a full-field transmission study of dislocations within the interior of a 400  $\mu$ m-thick synthetic diamond crystal, the second a magnified section topography study of a deformed SrTiO<sub>3</sub> sample and the third a full-field reflection study of a 120 nm BiFeO<sub>3</sub> thin film.

#### 2. The dark-field X-ray microscopy setup

Dark-field X-ray microscopy (Simons *et al.*, 2015) is conceptually similar to dark-field TEM. The experimental geometry and operational principle are shown in Fig. 1: monochromatic X-rays with wavelength  $\lambda$  illuminate the diffracting object. The sample goniometer comprises a base tilt,  $\mu$ , an  $\omega$  rotation stage, and two orthogonal tilts,  $\chi$  and  $\phi$ . The sample is oriented such that the Bragg condition is fulfilled, as defined by scattering vector **Q**, scattering angle  $2\theta$  and azimuthal angle  $\eta$ . An X-ray objective produces an inverted and magnified image in the detector/image plane. Furthermore, it acts as a band-pass filter in reciprocal space, selecting the  $\mathbf{Q}$  of a given diffraction spot, and thereby avoiding the problem of overlapping spots associated with other grains in a polycrystalline specimen.

The method's development has been motivated primarily by studies of polycrystalline samples. However, grains typically have to be aligned and studied one by one. For simplicity, in this article we shall assume the sample to be a single crystal. Furthermore, following current practice the objective will be a compound refractive lens (CRL) (Snigirev *et al.*, 1996) with *N* identical parabolic shaped lenses with a radius of curvature *R* and a distance between lenslet centres of *T*.

3D mapping can be achieved in two ways. Firstly, a line beam can be used to illuminate slices of the sample one at a time, and the 2D reconstructions are subsequently stacked. For some purposes this may be considered a magnified type of section topography, but the use of an X-ray objective implies a separation of angular and spatial degrees of freedom and as such adds additional advantages beyond the geometric magnification. Secondly, similarly to the topo-tomography approach mentioned above, one can use a full-field illumination and record projections from different viewing angles while rotating the sample about the scattering vector and subsequently using tomography-type algorithms to reconstruct the 3D volume.

Poulsen *et al.* (2017) provide a comprehensive description of the optical properties of the image plane, including expressions for the numerical aperture NA; the focal length  $f_N$ ; the relation between magnification  $\mathcal{M}$ , working distance  $d_1$ , and the distance between the lens exit and detector plane  $d_2$ ; and





Geometry of dark-field X-ray microscopy. The optical axis of the diffracted beam is defined by the centre of rotation of the sample goniometer, the centre of the objective and the point of normal incidence of the beam on the detector. **Q** is the scattering vector,  $2\theta$  is the scattering angle,  $\mu$ ,  $\chi$  and  $\phi$  are tilts, and  $\omega$  is a rotation around **Q**.  $d_1$  is the distance from the sample to the entry point of the objective,  $d_2$  the distance from the exit point of the objective to the detector and  $f_N$  the focal length of the objective. NT denotes the thickness of teh entire objective. The laboratory coordinate system  $(x_1, y_1, z_1)$  is shown.

the field of view, direct-space resolution and reciprocal-space resolution. It is shown how the local variation in tilt of the scattering vector (*i.e.* the local pole figure or mosaic spread) can be mapped by stepping the sample through two orthogonal tilts. The first is either the base tilt,  $\mu$ , or an equivalent rotation around  $y_l$  by a combination of tilts  $\chi$  and  $\phi$  – in both cases representing the 'rocking' of the sample in classical topography. The second is an orthogonal tilt, enabled by another combination of  $\chi$  and  $\phi$ . This represents the 'rolling' of the scattering vector. The axial strain can be measured by a longitudinal ( $\theta$ -2 $\theta$ ) scan, where 2 $\theta$  is varied by a combined translation and rotation of the objective and the detector.

Similarly to classical light microscopy, the hard X-ray microscope is associated with a 'Fourier plane', placed at a distance of  $f_N$  from the exit of the CRL (cf. Fig. 1). The intensity distribution in this back focal plane (BFP) is equivalent to the distribution in the Fraunhofer far-field limit. Poulsen et al. (2018) present a complementary description of the optical properties of the BFP. Here an alternative approach to mapping the local tilt and local axial strain is provided under the heading of local reciprocal-space mapping. By inserting an aperture in the BFP, a certain (small) region in reciprocal space is selected. The images acquired in the image plane will receive intensity only from real-space regions with the corresponding reciprocal-space state, for example only regions with a certain strain state. By translating the aperture within the BFP, the centre position of the region can be varied, e.g. the corresponding strain value can be scanned. Similarly to the operation of a transmission electron microscope (Williams & Carter, 2009), the possibility of combining local information in direct and reciprocal space is seen as a major asset of darkfield X-ray microscopy.

In the following we shall explore the microscope for mapping one axial and two off-diagonal strains around individual dislocations, corresponding to small variations in  $\phi$ ,  $\chi$  and  $2\theta$ . We will primarily be concerned with the contrast and resolution within a single image: algorithms for the generalization to 3D mapping will be presented elsewhere.

#### 3. Methodology

#### 3.1. Coordinate systems

Following the convention introduced by Poulsen *et al.* (2017), we define (for simplicity, we restrict the discussion in this article to the case  $\omega = \eta = 0$  and  $\mu = \theta$ ) a series of direct-space coordinate systems as displayed in Fig. 1. The laboratory coordinate system  $(\hat{x}_1, \hat{y}_1, \hat{z}_1)$  is defined with  $\hat{x}_1$  along the incident beam and  $\hat{z}_1$  vertical. As usual for imaging systems we will define the sample plane as a plane perpendicular to the optical axis, in this case the diffracted beam. This is spanned by  $(\hat{y}_s, \hat{z}_s)$  where  $\hat{y}_s = \hat{y}_1$ , and  $\hat{z}_s$  is inclined by the scattering angle  $2\theta$  with respect to  $\hat{z}_1$ . In the following, for brevity, we will use  $(x, y, z) = (x_s, y_s, z_s)$ .

In reciprocal space, we shall assume that the scattering vector probed is in the proximity of a reciprocal-lattice vector,  $\mathbf{Q}_0$ . Similarly to Poulsen *et al.* (2017), it is natural to introduce

three reciprocal-space coordinate systems. Their relationship is illustrated in Fig. 2 for  $\omega = 0$ . The *reference system*  $(\hat{q}_{\text{rock}}, \hat{q}_{\text{roll}}, \hat{q}_{\parallel})$  has  $\hat{q}_{\parallel}$  parallel to  $\mathbf{Q}_0$  and  $\hat{q}_{\text{roll}}$  parallel to the rolling direction and perpendicular to the vertical scattering plane. The *imaging system* is a parameterization which is colinear to the sample plane defined above. For  $\omega = 0$  this has coordinates  $(\hat{q}_{\text{rock'}}, \hat{q}_{\text{roll}}, \hat{q}_{2\theta})$ , with  $\hat{q}_{\text{rock'}}$  parallel to the optical axis. Finally, a coordinate system is introduced with its axis aligned with the direct-space laboratory coordinate system  $(\hat{q}_{1,x}, \hat{q}_{1,y}, \hat{q}_{1,z})$ .

#### 3.2. Weak beam contrast mechanism

In this article we will neglect effects due to (partial) coherence and assume that dynamical effects only take place within a sphere in reciprocal space around the lattice point,  $\mathbf{Q}_0$ , with radius  $q_{\rm dyn}$ . By definition, when probing parts of reciprocal space with  $|\mathbf{Q} - \mathbf{Q}_0| > q_{\rm dyn}$  kinematical scattering applies. We shall use the phrase 'weak beam contrast'.

We shall not be concerned with the symmetry of the unit cell, and reciprocal space and strain tensors both refer to a simple cubic system. Including crystallography is straightforward in principle, but the more elaborate equations make the treatment less transparent. Moreover, we will consider only the case of a synchrotron beam with an energy band  $\Delta E/E$  of order  $10^{-4}$  or less. Unless focusing optics are used the incoming beam will have a divergence of  $\Delta \zeta \simeq 0.1$  mrad or smaller. In comparison, the numerical aperture of the objective is much larger: NA  $\simeq 1$  mrad.

In the following we estimate the width of the intensity profile from a single straight dislocation within this weak beam contrast model. This estimate will be used for a simple comparison with experimental data and for discussing current and future use. For reasons of simplicity, we consider a fully





Definition of three coordinate systems in reciprocal space for  $\omega = 0$ : the reference system  $(\hat{q}_{\text{rock}}, \hat{q}_{\text{roll}}, \hat{q}_{\parallel})$ , the imaging coordinate system  $(\hat{q}_{\text{rock}'}, \hat{q}_{\text{roll}}, \hat{q}_{2\theta})$  and a system fixed with respect to the laboratory system  $(\hat{q}_{1,x}, \hat{q}_{1,y}, \hat{q}_{1,z})$ . All vectors except  $\mathbf{Q}_0$  are unit vectors and all vectors except  $\hat{q}_{1,y} = \hat{q}_{\text{roll}}$  are in the same plane. The sample plane normal is aligned with  $\hat{q}_{\text{rock}'}$ . In Section 3 a model is derived for scattering from a screw dislocation with the line and Burgers vector **b** aligned with  $\mathbf{Q}_0$ .

illuminated straight screw dislocation with Burgers vector **b** aligned with  $\mathbf{Q}_0$  and parallel to the *z* axis at x = y = 0 (*cf.* Fig. 2). In this case, when rotating around  $\mathbf{Q}_0$  the strain field and projections are invariant. In a classical dislocation model the non-zero strain components are

$$e_{zx} = -\frac{b}{2\pi} \frac{y}{x^2 + y^2}; \quad e_{zy} = \frac{b}{2\pi} \frac{x}{x^2 + y^2}.$$
 (1)

In general, the strain components  $e_{ij}$  associated with an isolated dislocation fall off as  $e_{ij} \simeq (b/2\pi)(1/r)$ , where *r* is the radial distance from the core of the dislocation.

For the simple cubic system and the case introduced above of a screw dislocation aligned with  $\mathbf{Q}_0$  (*cf.* Fig. 2) and  $\omega = 0$  we have

$$\frac{\Delta Q_{\text{rock}}}{|Q_0|} = -e_{zx}; \quad \frac{\Delta Q_{\text{roll}}}{|Q_0|} = -e_{zy}; \quad \frac{\Delta Q_{\parallel}}{|Q_0|} = -e_{zz}.$$
 (2)

#### 3.3. Mapping dislocations by magnified topography

It is shown by Poulsen *et al.* (2017) that in the imaging coordinate system (see Fig. 2) the resolution function is a Gaussian with the principal axis aligned with the coordinate axes and with widths (FWHM)

$$\Delta Q_{\text{rock}'} = \frac{|Q_0|}{2\cos(\theta)} \Delta \zeta, \qquad (3)$$

$$\Delta Q_{\rm roll} = \frac{|Q_0|}{2\sin(\theta)} \,\mathrm{NA},\tag{4}$$

$$\Delta Q_{2\theta} = \frac{|Q_0|}{2\tan(\theta)} \text{NA.}$$
(5)

This shows that  $\Delta Q_{\text{rock}'} \ll \Delta Q_{\text{roll}} \simeq \Delta Q_{2\theta}$  and the resolution function is in fact an oblate spheroid.

Comparing equations (1) and (2) with equations (4) and (5), it appears that, for experimentally relevant values of r, the intensities on the detector are the result of a 2D projection in reciprocal space: the objective's NA effectively integrates over directions  $\hat{q}_{2\theta}$  and  $\hat{q}_{roll}$ . In addition, the intensities are 1D projections in direct space, along the axis of the diffracted beam.

The resolution in the 'rocking direction' is in fact a convolution of the Darwin width of the sample and the divergence of the incoming beam. For simplicity, in equation (3) and throughout this article we shall neglect the Darwin width.

Next, let us consider the model system of Section 3.2. For  $\omega = 0$  we integrate over  $e_{zy}$ . The intensity distribution is then a function of only two variables  $I = I(y, e_{zx})$ . We can determine the path length along x for a given y and strain interval  $de_{zx}$  by inverting equation (1) and differentiating  $dx/de_{zx}$ . As a result

$$I(y, e_{zx}) \propto \int_{-\infty}^{\infty} f(y - y') \left| \int_{u_1}^{u_2} \frac{g(e_{zx} - u)}{u^2 \left( -\frac{b}{2\pi u y'} - 1 \right)^{1/2}} \, \mathrm{d}u \right| \, \mathrm{d}y', \quad (6)$$

with

$$u_1 = -\frac{b}{2\pi y'}; \quad u_2 = -\frac{by'}{2\pi [y'^2 + (T_c/2)^2]}.$$
 (7)

Here f(y) is the point spread function and  $g(e_{zx})$  is the resolution in  $e_{zx}$ . In the following we shall assume both to be Gaussian distributions.  $T_c$  is the thickness of the crystal in the direction of the diffracted beam.  $| \dots |$  symbolizes the absolute value.

Simulations of the intensity profile across a screw dislocation are shown in Fig. 3 using parameters relevant to the experiments presented later, including a point spread function f(y) with an FWHM of 180 nm, a strain resolution function  $g(e_{zx})$  with an FWHM of 0.02 mrad and a sample thickness of 400 µm. With increasing offset in rocking angle the width of the curves asymptotically approaches the spatial resolution, while the peak position in direct space, r, and strain (angular offset), e, approximately follow  $e = b/2\pi r$ .

For applications, a main challenge of any topography method is overlap of signal from dislocation lines. This effectively limits the approach in terms of dislocation density. It appears that in the weak beam contrast description the likelihood of overlap is determined by how far off the peak on the rocking curve one can go while still maintaining a contrast. The profiles shown in Fig. 3 are normalized. If not normalized, the amplitude of the profiles falls off rapidly with offset in rocking angle. Hence, signal-to-noise ratio becomes critical.

Another concern is the nature of the tails of the distributions f(y) and  $g(e_{zx})$ . If these tails are intense, such as in Lorentzian distributions, the contrast deteriorates. Hence, being able to design and characterize the resolution functions



Figure 3

Simulated intensity profile perpendicular to a screw dislocation with the offset in rocking angle in degrees as the parameter. All curves are normalized to 1. The abcissa represents distances in the sample in units of 100 nm. See text.

is important. Modifying  $g(e_{zx})$  can be achieved with an aperture in the BFP.

3.4. Mapping dislocations using an aperture in the back focal plane

Dark-field imaging is one of the basic modalities of a transmission electron microscope (Williams & Carter, 2009). By inserting an aperture in the BFP, one selects a certain region in reciprocal space and uses the diffracted signal within this region to generate contrast so as to image features within the sample such as dislocations. Poulsen *et al.* (2018) introduce the equivalent technique for hard X-ray microscopy. The relation between position ( $y_{\rm B}$ ,  $z_{\rm B}$ ) in the BFP, the angular offset in rocking angle  $\phi - \phi_0$  and reciprocal space is

$$q_{\rm rock} = \frac{\Delta Q_{\rm rock}}{|\mathbf{Q}_0|} = (\phi - \phi_0) - \frac{\cos(N\varphi)}{2\sin(\theta)f_N} z_{\rm B}\sin(\theta), \qquad (8)$$

$$q_{\rm roll} = \frac{\Delta Q_{\rm roll}}{|\mathbf{Q}_0|} = \frac{\cos(N\varphi)}{2\sin(\theta)f_N} y_{\rm B},\tag{9}$$

$$q_{\parallel} = \frac{\Delta Q_{\parallel}}{|\mathbf{Q}_{0}|} = \frac{\cos(N\varphi)}{2\sin(\theta)f_{N}} z_{\mathrm{B}}\cos(\theta), \tag{10}$$

with  $\varphi = (T/f)^{1/2}$  being a measure of the 'refractive power' of the lens, and  $f_N$  being the focal length. The last term in equation (8) and the  $\cos(\theta)$  factor in equation (10) originate in the fact that rocking the sample is a movement in a direction that is at an angle of  $\theta$  with the optical axis (the direction of the diffracted beam).

Unfortunately, if the aperture gap D is smaller than or comparable to the diffraction limit  $\lambda$ /NA, the spatial resolution in the imaging plane will deteriorate. On the other hand, using wavefront propagation Poulsen et al. (2018) demonstrated that the aperture will not influence the spatial resolution if the gap is sufficiently large. For a specific application introduced below, the minimum gap is 80 µm. In order to provide a high resolution both in reciprocal space and in direct space, we therefore propose to move a square aperture with a sufficiently large gap in a regular 2D grid within the BFP and to regain reciprocal-space resolution by a deconvolution procedure as follows: let the positions of the centre of the slit be  $(y_{\rm B}, z_{\rm B}) = D/M \times (m, n)$ , with m = -M, -M + 1, ..., Mand  $n = -M, -M + 1, \dots, M$ . For fixed (m, n), for fixed rocking angle  $\phi$  and for a given pixel on the detector, let the set of intensities measured in this detector pixel be  $S_{m,n}$ .

Now, consider the intensities  $I_{m,n}$  for an aperture of size D/M, in the hypothetical case that the diffraction limit can be neglected. Moreover, assume the diffracting object is bounded such that there is no diffracted intensity outside the grid. Then, in the first quadrant we have, for  $0 \le m < M$  and  $0 \le n < N$ ,

$$I_{m,n} = S_{m,n} - S_{m+1,n} - S_{m,n+1} + S_{m+1,n+1}.$$
 (11)

This relationship is illustrated in Fig 4. For the other quadrants similar expressions can be established. Hence, using this simple difference equation we can generate high-resolution q maps.

Poulsen *et al.* (2018) also found that the FWHM of the resolution function in the BFP can be  $\Delta Q/|\mathbf{Q}_0| = 4 \times 10^{-5}$  or better in all directions, which is substantially smaller than the angular range of the diffracted beam. We conclude that by placing an aperture in the BFP we can generate a 5D data set. Hence, we can associate each detector point with a reciprocal-space map. Then the only remaining integration is in the thickness direction in real space.

A significant simplification arises if we use the formalism of elasticity theory. Then each point (x, y, z) in the sample is associated with one point in reciprocal space corresponding to the three strain components:  $(e_{zx}, e_{zy}, e_{zz})$ . Let the recorded intensities be  $I_{m,n}(\mathbf{q}, y_d, z_d)$  with  $(y_d, z_d)$  being the detector coordinates,  $\mathbf{q} = (q_{\text{rock}}, q_{\text{roll}}, q_{\parallel})$  and strain vector  $\mathbf{e} = (e_{zx}, e_{zy}, e_{zz})$ . Then for  $\omega = 0$  we have

$$I_{m,n}(\mathbf{q}, y_{\mathrm{d}}, z_{\mathrm{d}}) \propto \int \int \int dx_{\mathrm{s}} du \, dv f(y_{\mathrm{d}} - u, z_{\mathrm{d}} - v) \\ \times \int d^{3} \mathbf{q}' g[\mathbf{e}(x, u/\mathcal{M}, v/\mathcal{M}) - \mathbf{q}'].$$
(12)

Here  $\mathcal{M}$  is the magnification in the X-ray lens and f is the detector point spread function. For sufficiently small steps D/M in the deconvolution algorithm [equation (11)] g will approximate the superior reciprocal-space resolution function in the BFP.

For the one-dislocation model introduced in Section 3.2 this implies that the intensity profiles across a dislocation line in general become substantially narrower than those displayed in Fig. 3. For small D/M the point spread function dominates. This will enable studies of samples with higher dislocation densities. Moreover, one may use the distributions to separate dislocations that are overlapping and inseparable in the greyscale images. We anticipate this enhanced contrast to be useful in several other ways as well, including for identifying Burgers vectors, and for guiding and validating models of dislocation interactions.



Figure 4

Illustration of the difference equation [equation (11)] used to derive the intensity  $I_{m,n}$  from the integrated intensities  $S_{m,n}$ ,  $S_{m+1,n}$ ,  $S_{m,n+1}$  and  $S_{m+1,n+1}$ . For high numbers of *m* and *n* the intensities *I* are assumed to be vanishing. See text.

With respect to implementation, it may also be possible to transfer additional TEM modalities. In particular, annular dark-field imaging is a candidate for fast 3D mapping of dislocations. Blocking the central beam may be an elegant way to remove spurious effects due to dynamical diffraction.

#### 4. Experimental demonstrations

To illustrate the potential and challenges of our approach, we report on the results from three different types of use. Three samples were studied at beamline ID06 at the ESRF over two beamtimes and under slightly different configurations (as the beamline instrumentation evolved during this period).

In all cases, an Si(111) double monochromator was used to generate a beam with an energy bandwidth of  $\sigma_e = 0.6 \times 10^{-4}$ (r.m.s.). The goniometer with all relevant degrees of freedom (cf. Fig. 1) is placed 58 m from the source. Pre-condensing is performed with a transfocator (Vaughan et al., 2011) positioned at a distance of 38.7 m from the source. For section topography, a 1D condenser was used to define a horizontal line beam. Otherwise, a slit defined the dimensions of the beam impinging on the sample. Two detectors were in use: firstly, a near-field camera, placed close to the sample, which can provide classical topographs and topo-tomograms without the magnification by the X-ray objective; secondly, a far-field camera placed at a distance of  $\sim$ 5.9 m for imaging the magnified beam in the image plane of the microscope. Both detectors were FRELON 2k × 2k CCD cameras (Labiche et al., 2007), which are coupled by microscope optics to a LAG scintillator screen (Douissard et al., 2012). The objective comprised N identical parabolically shaped Be lenses with a radius of curvature  $R = 50 \,\mu\text{m}$  and thickness T. A square slit with adjustable gaps and offsets was placed in the BFP. The surface normals of all detectors and slits were aligned to be parallel to the optical axis. The near-field camera and the

aperture in the BFP could be translated in and out of the diffracted beam.

#### 4.1. Transmission experiment

The sample was an artificially grown diamond plate, type IIa, with a thickness of 400  $\mu$ m (see Burns *et al.*, 2009). It was mounted in a transmission Laue geometry. The 17 keV incident beam had a divergence (FWHM) of 0.04 mrad. For work with the diffraction microscope the beam was confined to dimensions of 0.3 × 0.3 mm. With N = 72 and T = 2 mm, the focal length of the objective was  $f_N = 0.245$  m. The effective pixel sizes of the near- and far-field detector were 0.62 and 1.4  $\mu$ m, respectively. The magnification by the X-ray objective was measured to be  $\mathcal{M} = 16.2$ , implying a numerical aperture of NA = 0.643 mrad and an effective pixel size of 93 nm. The detector was then binned 2 × 2. Using equations (3)–(5) the FWHMs of the reciprocal-space resolution function in the three principal directions become  $(\Delta q'_{\rm rock}, \Delta q_{\rm roll}, \Delta q_{2\theta}) = (0.00062 \text{ Å}^{-1}, 0.0055 \text{ Å}^{-1}, 0.0055 \text{ Å}^{-1}).$ 

An in-plane {111} reflection was used for the study. The lengths of the diffraction vector and Burgers vector are  $|\mathbf{Q}_0| = 3.051 \text{ Å}^{-1}$  and  $|\mathbf{b}| = 2.522 \text{ Å}$ , respectively. Using the formalism of Als-Nielsen & McMorrow (2011), the corresponding Pendellösung length and Darwin width are  $\Lambda_g = 35 \,\mu\text{m}$  and  $w_g^{\theta} = 0.0119 \,\text{mrad}$  (FWHM), respectively. Hence, the incoming beam divergence dominates the Darwin width. The data set involved 36  $\omega$  projections over a range of 360°. For each projection, images were acquired in a 31 × 31 grid in rocking angle  $\mu$  (with steps of 0.0016°) and  $2\theta$  (steps of 0.0032°). Exposure times were 1 s.

Fig. 5 shows an image from the near-field detector and the corresponding dark-field image from the diffraction microscope. The latter is inverted for ease of comparison. Three dislocations are present in the latter, all exhibiting kinks, as well as several scratches on the surface. The difference in field

> of view is evident, as is the fact that the objective magnifies the image without visible distortions.

> Fig. 6 shows the diffracted signal as a function of rocking angle from a specific location in the microscope image. It appears that the signal is corrupted by dynamical diffraction effects until at least  $\delta \mu = \pm 0.002^{\circ}$ . The signal-to-noise ratio allows useful observations out to  $\delta\mu \simeq \pm 0.008^{\circ}$ , corresponding to a transverse strain of  $\pm 1.4 \times 10^{-4}$ . Similar plots of the intensity profile in the radial direction (obtained by a simultaneous translation in  $\mu$  and  $2\theta$  by  $\delta\mu = \frac{1}{2}\Delta 2\theta$ ) – also known as the 'longitudinal direction' - showed a very similar sensitivity. Hence, both 'rocking' and 'longitudinal' contrast are validated. As expected, no contrast was detectable in the rolling and  $2\theta$  directions, owing to the



#### Figure 5

Projection images of a large single-crystal diamond in the transmission experiment. (Left) Nearfield detector image with no X-ray objective and (right) corresponding dark-field image acquired with the diffraction microscope, both for  $\mu - \mu_0 = 0.002^\circ$ . The magnification of the microscope is  $\mathcal{M} = 16.2$ . The direction of the rotation axis is marked by an arrow.  $\hat{q}_{2\theta}$  and  $\hat{q}_{roll}$  are parallel to the x and y axes of these subfigures, respectively. convolution of the diffracted signal with the numerical aperture of the objective.

In Fig. 7 (left) two diffraction images are overlaid, corresponding to the left and right of the Bragg peak on the rocking



#### Figure 6

Zoom of data from the transmission experiment, each image showing one screw dislocation (left) attached to a triangular area associated with a stacking fault. The variation with rocking angle  $\delta \mu$  is shown. The line plots represent the integrated intensity as a function of distance perpendicular to the dislocation line, as marked by the five-pixel-thick black lines. The line plots are normalized to maximum intensity. The red lines indicate the interpolated position of the dislocation line.  $\hat{q}_{2\theta}$  and  $\hat{q}_{roll}$  are parallel to the *x* and *y* axes of these subfigures, respectively.

curve. As anticipated, the signal is antisymmetric with respect to the diffraction lines. Line profiles of the intensity across the dislocation lines reveal that a centre line between the purple and green curves can be established with high accuracy, 50 nm

or better. Compared with the corresponding signal from the near-field camera (see Fig. 7 right), the contrast and resolution of the dark-field microscopy setup are clearly better. However, the resulting width of the dislocation line is approximately 1.5  $\mu$ m FWHM. In comparison, the simple kinematical model of Section 3.2 predicts a width of ~200 nm (*cf.* Fig. 3).

To estimate whether depth of field could be the cause of this effect, we note that a given incoming ray traversing through the strain field of a dislocation can be scattered in different directions. When the dislocation is in the sample plane, these diverging rays are all collected in the image plane. If the dislocation is displaced by, for example, 100 µm along the optical axis, a geometrical optics expression, equation (13) of Simons et al. (2017), predicts a diffraction-limited (real-space) resolution with an FWHM of 100 nm for a strain range of  $\pm 1.4 \times 10^{-4}$ . Hence, depth of field cannot be the cause.

The dominant cause of discrepancy is instead considered to be alignment of the microscope, which was problematic at the time owing to the *ad hoc* character of the setup.

# 4.2. Magnified section topography experiment

Within the weak beam regime one may reduce the likelihood of overlap of dislocations in the images by narrowing the incident beam in the vertical direction (see Fig. 3). By introducing a condenser we can furthermore improve the signal-to-noise ratio, at the expense of an increased divergence. In principle, one can adjust the height of the incoming beam to match the spatial resolution. 3D mapping can then be performed layer by layer. However, identifying points is more difficult than identifying lines, and 1D condensers providing a micrometre-sized beam tend to be more efficient than those producing a nanometre-sized beam. Hence, it may be optimal to operate

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Figure 7

Diffraction images from the same region acquired with the diffraction microscope (left) and the near-field camera (right). In both cases two images are overlaid, a purple one and a green one representing offsets of the rocking angle  $\mu$  by + 0.002 and -0.002°, respectively. Shown in the middle are line plots of the green images representing the intensity distribution perpendicular to the dislocation line.

with an incoming box beam having a large aspect ratio. We shall use the term 'magnified section topography' for this setting.

In this experiment, the sample was a wedge-shaped piece of SrTiO<sub>3</sub>, where surfaces had been polished mechanically. It was mounted in a transmission Laue geometry, using a {110} reflection for the study. The 15.6 keV beam was condensed by a CRL with 55 1D Be lenslets to generate a beam (FWHM) of size  $4.2 \times 300 \,\mu$ m. The objective configuration was in this case N = 45,  $T = 1.6 \,\text{mm}$ , leading to a focal length of  $f_N = 0.406 \,\text{m}$ . The measured X-ray magnification was 12.32 and consequently the numerical aperture had an r.m.s. width of  $\sigma_a = 0.24 \,\text{mrad}$ . The far-field detector had an effective pixel size of

122 nm. A rocking scan was made over a range of  $0.5^{\circ}$ , with 70 steps and exposure times of 1 s.

Fig. 8 shows a raw image. The top point of the wedge is far to the left of this image. Generally speaking, the weak beam scattering signal is confined to two regions, adjacent to the two external boundaries (top and bottom in the figure). The dislocation density is too high in these regions to resolve the individual dislocations. We speculate that these have formed during polishing. As shown in the figure, at a certain distance from the top of the wedge, dislocations are created that thread the thickness and bridge the gap between the two surface layers. The intensity profile across one of these vertical lines is shown in Fig. 9. As shown in the figure, the profile is well described by a Lorentzian model. A fit to this model results in a half-width at half-maximum (HWHM) of 210 nm. In Fig. 8 in the vicinity of the prominent vertical dislocations a network of



Figure 8

A raw image from the magnified section topography study of an SrTiO<sub>3</sub> wedge sample where regions near surfaces (top and bottom) are deformed owing to mechanical polishing. In the middle there are threading dislocations. The offset in rocking angle is 0.5 mrad. One of the dislocations is marked by an arrow.  $\hat{q}_{roll}$  and  $\hat{q}_{2\theta}$  are parallel to the *x* and *y* axes of the figure, respectively.



Figure 9

Intensity profile across the dislocation marked by an arrow in Fig. 8 (dots) and corresponding fit to a Lorentzian (line). The fitted HWHM value is 210 nm.

other dislocations pointing in near random directions is seen. Their line widths are in some cases below 200 nm, but the statistics are poor. The 200 nm is slightly larger than the spatial resolution of the instrument.

#### 4.3. Reflection experiment

Mapping individual dislocations is of great interest also for films and buried layers. Often these have to be studied in a reflection geometry, as the X-rays cannot penetrate the substrate. The reflection geometry implies a parallax effect in the vertical direction and 3D mapping requires special algorithms, e.g. laminography (Hänscke et al., 2012). To illustrate the potential of hard X-ray microscopy for such samples, we have studied misfit dislocations in BiFeO<sub>3</sub> thin films. First results were presented by Simons, Jakobsen et al. (2018). In short, individual dislocations are identified, and their axial strain field characterized by means of a ' $\theta$ -2 $\theta$  scan': a combined translation and rotation of the sample, the objective and the far-field detector. Here we report on additional work, where we illustrate the reciprocal-space mapping introduced in Section 3.4 by means of translating an aperture in the BFP. The ultimate aim for this type of study is to repeat the reciprocal-space mapping for a set of  $\omega$  projection angles in order to reconstruct the strain field for each voxel in the sample.

Addressing this challenge is an exercise in vector tomography (Schuster, 2008) and is outside the scope of this article. Here a simple data analysis is presented for the case of one projection.

The sample was a 120 nm-thick film of (001)-oriented BiFeO<sub>3</sub>, grown via pulsed laser deposition on an SrRuO<sub>3</sub> electrode layer and a  $\langle 110 \rangle$ -oriented DyScO<sub>3</sub> single-crystalline substrate. This was mounted for a reflection study on the (002) reflection at  $2\theta = 22.6^{\circ}$ . In this case the 15.6 keV beam from the transfocator was only moderated by a slit close to the sample. The objective and detector configuration were identical to those of Section 4.2. The aperture in the BFP had a square opening of 80 µm, which is slightly larger than the size of the beam in that plane. Within the approach of Section 3.4 this aperture was translated in a 2D grid with a step size of 30 µm. At each position a rocking scan was made with a step size of  $0.001^{\circ}$  and with exposure times of 2 s.

The signal was deconvoluted according to equation (11), and each point in the sample plane was associated with a reciprocal-space map. The voxel size of this map is  $\Delta Q/|Q| = (1.7 \times 10^{-5}, 1.6 \times 10^{-4}, 1.6 \times 10^{-4})$  in the rock', roll and  $2\theta$  directions, respectively.

Zooming in on one dislocation, we illustrate in Fig. 10 the richness of the results obtained. To the left is shown the result with no aperture in the BFP for two offsets in rocking angle. These images represent an integration over  $e_{zy}$  and  $e_{zz}$  at the given  $e_{zx}$  (as determined by the position of the rocking angle  $\phi$ ). The remainder of the subplots are corresponding results based on the aperture scan. For each point in the detector plane a Gaussian fit was made to the intensity profile arising from scanning the aperture horizontally. Using equation (9) this is converted into a relative shift  $q_{\rm roll}$ . The fitted centre position and width (FWHM) are shown in columns 2 and 3, respectively. In columns 4 and 5 are shown the results of an analogous fit to the intensity profile arising from scanning the aperture vertically. Using equation (10) this is converted into a relative shift  $q_{\parallel}$ . Apart from a change in sign these figures can be directly interpreted as maps of the centre and the width of the strain components  $e_{zy}$  and  $e_{zz}$  – for the given value of  $e_{zx}$ [cf. equation (2)].

The rocking profiles (not shown) exhibit a clear asymmetry, analogous to that shown in Fig. 6. The second column of Fig. 10 reveals that the rolling profiles have a similar left–right asymmetry. Near the dislocation core the profile has a dip in the centre, evident as a large increase in the FWHM of the one-peak fit (third column). In contrast there is no noticeable variation in the longitudinal direction (columns 4 and 5).



#### Figure 10

Images of a dislocation in a BiFeO<sub>3</sub> film acquired at an offset in rocking angle from the main peak of  $\phi = 0.01^{\circ}$  (top row) and  $\phi = 0.015^{\circ}$  (bottom row). The contrast is set differently in the two rows. First column: no aperture in the BFP; red is maximum intensity, blue is background.  $\hat{q}_{roll}$  and  $\hat{q}_{2\theta}$  are parallel to the *x* and *y* axes of these subfigures, respectively. The other four columns: results from scanning an aperture of fixed size in the BFP. For each pixel on the detector, Gaussian-type fits were made to the profile in the rolling and longitudinal directions. Shown are the centre-of-mass positions and the FWHM in units of  $\Delta Q/|Q_0|$ . The widths are determined by equations (9) and (10), respectively. The unit on the axes is  $\mu$ m and refers to the detector plane.

These findings are all consistent with the response from the strain field from a single dislocation with the Burgers vector pointing in the direction of the surface normal.

#### 5. Discussion

Dark-field microscopy is fundamentally different from classical X-ray topography, as rays emerging in various directions from one point in the sample plane are focused onto a spot in the image plane, rather than leading to a divergent diffracted beam. This implies that the detector can be placed many metres away and that the space around the sample is limited by the objective, not the detector. Moreover, the high spatial resolution allows one to visualize the core of the strain field. This simultaneously enables the dislocations to appear as thin lines and scattering to be sufficiently offset from the Bragg peak that weak beam conditions apply. Below we first present the perceived main limitations of the technique and discuss options to overcome these. Next we briefly outline the scientific perspective.

Dynamical diffraction effects. The 'weak beam' condition presented strongly simplifies the data analysis and interpretation. In practice, it is likely that dynamical or coherent effects need to be considered in some cases. A treatment of dynamical scattering in the context of X-ray topography can be found in the work of, for example, Gronkowski & Harasimowicz (1989) and Gronkowski (1991). However, as mentioned previously, the geometry of data acquisition is fundamentally different in a microscope. A dynamical treatment of the scattering of a dislocation line in the context of a microscope exists for TEM (Hirsch *et al.*, 1960), but to the knowledge of the authors has yet to be generalized to X-ray microscopy. In a heuristic manner, with dark-field microscopy we attempt to overcome the issue with dynamical effects in two ways:

(1) By improving both the spatial and angular resolution it becomes possible to probe parts of reciprocal space which are further from  $q_{dyn}$ .

(2) By combining projection data from a number of viewing angles we anticipate that 'dynamical effects can be integrated out'. Similar strategies have led the electron microscopy community to apply annular dark-field imaging for providing accurate crystallographic data (Nellist & Pennycook, 2000).

*Spatial resolution.* The spatial resolution sets an upper limit on the density of dislocations that can be resolved. With increasing spatial resolution, one can monitor the strain and orientation fields closer to the core. At the same time, dynamical diffraction effects become smaller as one is probing parts of reciprocal space that are further away from the Bragg peak. In practice, the limitation of the technique is currently set by aberrations caused by the lens manufacture and by signal-to-noise considerations. Moreover, for the studies presented here – based on *ad hoc* instrumentation – alignment is an issue. With the possibility of providing a reciprocal-space map for each voxel in the sample (*cf.* Section 3.4), overlap of the diffraction signals from dislocation lines can be handled. It is our understanding that there is no fundamental physics prohibiting a substantial increase in the spatial resolution of a dark-field microscope. With ideal CRL optics hard X-ray beams may be focused to spot sizes below 10 nm (Schroer & Lengeler, 2005). Using zone plates as objectives, at X-ray energies below 15 keV, bright-field microscopes are in operation with resolutions at 20 nm. For work at higher X-ray energies, there has recently been much progress with multilayer Laue lenses, which seem to promise imaging with superior numerical apertures and much reduced aberrations (Morgan *et al.*, 2015). Finally, the next generation of synchrotron sources will be 10–100 times more brilliant than the current sources (Eriksson *et al.*, 2014). This will benefit both spatial resolution (via improved signal-to-noise ratio) and time resolution.

Probing only one diffraction vector. As for any other diffraction technique, the contrast in visualizing the dislocations is proportional to  $\mathbf{Q} \cdot \mathbf{b}$ . Dislocations with a Burgers vector nearly perpendicular to the  $\omega$  rotation axis are therefore invisible. In order to map all dislocations and/or to determine all components of the strain tensor one has to combine 3D maps acquired on several reflections.

Scientific outlook. The higher resolution in three dimensions offers new perspectives on dislocation geometry, including measurements of distances and dislocation curvatures (and the balance of line tension by local stresses). This may be relevant for models of dislocation dynamics and the visualization of dislocations, for example, under indentations. With respect to dynamical diffraction effects, we recall that extinction lengths for 30 keV X-rays are about 100 times larger than the corresponding extinction lengths for 200 keV electrons. This points to high-resolution studies of dislocation dynamics in foils at least 10 µm thick.

Studies of dislocation structures within grains or domains are facilitated by the fact that dark-field microscopy is easy to integrate with coarse-scale grain mapping techniques such as 3D X-ray diffraction (Poulsen & Fu, 2003; Poulsen, 2012; Hefferan *et al.*, 2012) and diffraction contrast tomography (King *et al.*, 2008; Ludwig *et al.*, 2009).

#### 6. Conclusion

We have demonstrated an X-ray microscopy approach to characterizing individual dislocations in bulk specimens. The method combines high penetration power, a data acquisition time for 3D maps of minutes, and the possibility to study local internal regions by magnifying the images. In the weak beam contrast description provided, the likelihood of overlap is determined by how far off the peak on the rocking curve one can go while still maintaining a contrast. The spatial resolution (HWHM) in this proof-of-concept work is 200 nm. The limitation is the quality of the focusing optics, the signal-tonoise ratio and the alignment of the instrument. With improved X-ray sources and optics this opens the door to studies with a substantially higher spatial resolution. The high resolution allows studies of samples with higher densities of dislocations, and at the same time it enables one to probe the material at rocking angles with a large offset from the main peak, where the weak beam condition is fulfilled.

The method can be extended to mapping of the  $e_{zx}$ ,  $e_{zy}$  and  $e_{zz}$  fields by scanning a fixed gap aperture in the BFP of the objective and by rocking the sample.

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