Weak-beam dark-field electron tomography of dislocations in GaN

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Abstract. By combining weak-beam dark-field imaging with tomography, we have been able to reconstruct the three-dimensional structure of dislocation arrays in GaN. With a mixture of both threading and in-plane dislocations, owing to plastic relaxation of the film, we look at how well each dislocation is reconstructed and what limits are imposed by way of dislocation density and material anisotropy.

1. Introduction
Tomographic imaging in the electron microscope appears to be enjoying a renaissance of late. Two key factors seem to be responsible for this: the ability to quantitatively measure electron scattering & the availability of fast computer algorithms able to reconstruct useful volumes of material (aided by a commensurate drive towards nano-scale objects). The scattering mechanism itself can be anything provided that the signal measured obeys the projection requirement, which states that the signal must be a monotonic function of both the sample thickness and the property responsible. For example, in STEM-HAADF tomography the intensity increases both with thickness and atomic number and this provides reconstructions in which regions can be distinguished by atomic number [1].

At first hand, diffraction contrast does not appear to fulfill the projection requirement. Intensity oscillations caused by thickness gradations are a well known example [2]. However, diffraction contrast of dislocations will obey the projection requirement if they do not overlap significantly and the contrast be made small and local to the dislocation core. In the weak-beam dark-field imaging technique this is indeed the case provided the weak-beam diffraction condition can be maintained and is constant over the whole tilt range. This paper looks at how we have achieved this.

Three-dimensional dislocation analysis is not new. As early as 1965, Lang obtained three-dimensional information of dislocations with stereographic pairs of x-ray topographs [3]. However, stereography has a very limited depth resolution, so it was not until 2001 that full three-dimensional reconstructions were obtained by Ludwig and co-workers [4]. They were able to reconstruct dislocations in bulk, millimeter-sized pieces of synthetic diamond using a tilt series of Lang-topographs, so called topo-tomography, over a 360° tilt range. They were able to resolve individual dislocations provided they were spaced by more than a few microns, giving a practical limit of about $10^6$ dislocations cm$^{-2}$ for their method. Tomography of higher densities of dislocations requires a different approach for which the transmission electron microscope seems already perfectly equipped.
2. Theory
Dislocations of Burgers vector $\mathbf{b}$ appear in a dark-field (DF) image formed with diffracted beam, $\mathbf{g}$, if the following condition is obeyed: $\mathbf{g} \cdot \mathbf{b} > 0$. Tilting the crystal to put $\mathbf{g}$ out of Bragg condition, constrains the dislocation contrast to regions adjacent to the dislocation, owing to counter-rotation of the crystal lattice. The position of this DF intensity maximum occurs for the electron ray that threads the position $r_0$, where the effective deviation parameter goes through zero [5]:

$$s_{eff} \approx s_g + \mathbf{g} \cdot \left[ \frac{d\mathbf{R}}{dz} \right]_{r_0} = 0$$

where $z$ describes the direction of the electron beam and $\mathbf{R}$ is the displacement field around the dislocation. For a known displacement field, the position of $r_0$ can be found exactly. The distance away from the core for which equation (1) is obeyed is typically several nanometres.

During the tilt series acquisition, over a tilt range $\pm \theta_{max}$, the whole sample has to remain in focus. With a small objective aperture (~1 mrad), the Rayleigh resolution of the weak-beam dark-field (WBDF) image is quite large ($d_R = 0.6 \lambda / ~1nm$). The distance over which the image remains in focus i.e. the depth of field, is consequently also large ($=d_R / ~1\mu m$). Thus provided that the features of interest in the tilt series do not exceed a lateral distance of $\cot(\theta_{max})$, then the tomogram will suffer no resolution loss.

3. Experiment
We used hetero-epitaxial GaN grown on sapphire with high dislocation densities $\sim 10^9 - 10^{10} \text{cm}^{-2}$ caused by the large lattice mismatch (~14%). These samples usually have only threading dislocations with line vector along [0001]. However, we studied GaN epilayers with a 1µm GaN layer doped with Mg to make it p-type, for which cracking occurred. This led to both threading and in-plane dislocations to image. Samples were prepared by mechanical polishing and ion-milling in plan view with the [0001] axis orientated near the optic axis.

WBDF images were taken in a modified tilt-rotate specimen holder. By removing some of the material at the sides of the holder, we were able to get a ±60° tilt range in a standard 300kV TEM (Philips CM30). With [0001] near the optic axis, it was easy to find a weak-beam condition $\mathbf{g} / \mathbf{n}$ such that $n \approx 5$ ($s_g = 2.9 \times 10^{13} \text{Å}^{-1}$) for the reciprocal lattice vector $\mathbf{g} = 11-20$. Earlier work showed that the vast majority of the dislocations in this sample had a Burgers vector component along <11-20>.

WBDF images were taken with electron sensitive film on a plate camera and at 13500× magnification. Twenty five images at 5° tilt intervals over a 120° tilt range were taken with lenses kept at constant excitation to prevent changes in the image magnification and rotation. Refocusing was done manually using the eucentric specimen height control. The excitation error was kept constant for the tilt series. Negatives were digitised with a sample rate corresponding to 1.1nm per pixel. Images in the tilt series were then filtered and aligned to form the raw tilt series.

Tomographic reconstructions were performed with IDL using back-projections and sequentially iterated reconstruction technique (SIRT) to 10 iterations per slice, so that the total reconstruction volume corresponded to 600×600×150 nm³.

4. Results and Discussion
Figure 1(a) shows one of the 25 WBDF images taken in the tilt series. Evident in this image is a crack (running from the top to the lower right), which has, associated with it, a dense bundle of dislocations. Coming off this crack, almost horizontally are several in-plane dislocations that remained visible throughout the tilt series. Also evident are thickness fringes and bend contours that dominate the image contrast towards the thinnest part of the specimen.

Removal of the broader contrast variations was achieved by subtracting off a repeatedly smoothed version of the original image as shown in figure 1(b). This filter emphasized the dislocations very effectively including previously weak threading dislocations. However, it was not completely successful in removing the thickness fringes.
Two voxel projections of the reconstructed dislocation array are shown in figure 2. The entire reconstruction has a dusty appearance owing to the algorithm trying to reconstruct the rather mobile thickness fringes in the post-filtered images. It is particularly noticeable towards the bottom of the reconstructions where the fringes are strongest (figure 2a).

Reconstruction fidelity varied for the dislocations. Threading dislocations in the film were poorly reconstructed, having a `string-of-pearls’ appearance that faded towards the bottom of the sample. This appearance can be explained by a combination of intensity oscillations associated with inclined extended defects and anomalous absorption of the beam [2]. In-plane dislocations were very well reconstructed owing to their near constant visibility over the whole tilt range. Particularly evident are the curved dislocation segments near the crack labeled T in figure 2b. These dislocations are in fact threading dislocations that turn-over in response to the shear-stress field around the crack-tip bundle of dislocations. These dislocations glide away from the bundle forming a screw segment. Evidence of glide is shown by the dislocation that glides into our field of view from a neighbouring crack (see

Figure 1. A WBDF image at the g/4g condition with g=(11-20) taken at a tilt of +30° (a). After removing a smoothed version of the boxed area, the thin lines of the dislocations remain, but so does some of the contrast of the thickness fringes (b).

Figure 2. A perspective view of the whole reconstructed volume (a). Threading dislocations show a ‘string-of-pearls’ appearance (TD) with reduced visibility towards the bottom and an in-plane dislocation (G) bends up to intersect the top surface. Towards back of the volume (b), in-plane dislocations show evidence of turn-over (T), dislocation interaction (I) and kinking (K).
G indicated in figure 2a). These dislocations also show evidence of collisions with threading dislocations and kink-formation (I and K respectively in figure 2b). The images of in-plane dislocations were reconstructed to be elongated in the direction of the specimen normal. This was in excess of the factor 1.5 expected for this limited tilt range. This may be explained by the anisotropy of GaN. The stationary point $r_0$ moves as the sample is tilted with the following dependence on tilt angle, [7]:

$$r_0 = \frac{g \cdot b}{2 \pi s_{\text{eff}}} \frac{\sqrt{c_{66}/c_{44}}}{\left(1 + \left(\frac{c_{66}}{c_{44}} - 1\right) \sin^2 \theta\right)}$$  \hspace{1cm} (2)

where the $c$'s are the elastic stiffness constants of GaN ($c_{44}=24$ GPa, $c_{66}=83$ GPa). For the tilt range used here ($\pm 60^\circ$) two core-to-peak distances are found: 6.3nm (specimen tilt, $=60^\circ$) and 10.2nm ($=0^\circ$) for an in-plane screw dislocation (the ratio $c_{66}/c_{44} \approx 3.5$ GaN c.f. $c_{66}/c_{44} \approx 1700$ for graphite [9]). So for no more than a three-fold change in peak position, the ratio should be $c_{66}/c_{44} < 3$, as a general rule.

The turn-over of the threading dislocations near the crack tip dislocation bundle is not unexpected. This material had numerous elongated cracks which appeared with long <1-100> oriented dislocation bundles that formed ~1µm bands. These regions have a strong shear stress gradient both along the growth direction and in the plane of the film. The resolved shear stress forces dislocations with outward Burgers vectors to glide away from the stress source. The glide occurred only when the sample was treated by a rapid thermal anneal (untreated samples did not show such in-plane dislocations).

Our reconstruction was not able to resolve dislocations that formed the crack tip bundle. These dislocations appeared to have spacing of about ten nanometres, whereas dislocations spaced by about 100nm were resolved. Therefore, WBDF tomography looks set to be able to resolve dislocations with densities approaching $10^{12}$ cm$^{-2}$, six orders of magnitude greater than the previous x-ray technique.

5. Conclusions
In conclusion we have successfully reconstructed an array of dislocations in GaN in three dimensions with varying degrees of success. Dislocations that appear vertical in the film were not as well resolved as those in the plane of the sample. Artefacts in the reconstruction suggest that the degree of material anisotropy will govern the success of the reconstruction of other materials.

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