Quantitative High-Resolution TEM/STEM and Diffraction

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Charge density within a unit cell of GaN imaged withsub-Ångstrøm resolution by differential phase contrast microscopy

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Gallium nitride (GaN) is a highly interesting material for optoelectronics (high power LEDs and laser diodes) and actuators. The possibility to form ternary and quaternary alloys with InN and/or AlN allows a modification of the band gap and to tune the emission wavelength continuously from infrared to ultraviolet light. GaN also posesses a spontaneous polarization which causes local fields up to 250 MV/m between the intrinsic sheet charge densities. When strained, a piezoelectric polarization builds up in addition, which causes a tilt of the band structure across quantum wells. This tilt is believed to be responsible at least in part for a reduced performance due to the quantum confined Stark effect (QCSE) for laser diodes emitting in the green spectral range. For a direct measurement of local electric fields, we use the differential phase contrast (DPC) technique (see below) to monitor in STEM mode the minute deflections of the central diffraction disk when the beam passes through a region with a static field. To achieve the resolution needed we used a FEI Titan cubed 60-300kV, equipped with a high brightness gun, monochromator, an image C_s corrector and DCOR probe corrector with a convergence semi angle of about 21 mrad and a probe size of about 0.8Å. The detector used was a dedicated four quadrants solid state detector. The differential phase contrast technique was originally suggested by Rose [1] and later used in large extent to investigate micromagnetic structures in a STEM [2-7]. Recently, its capability to image electrostatic fields has been demonstrated, first on a mesoscopic (nm) scale [8], shortly followed on a microscopic (sub-nm) scale [9]. The specimen is scanned with a finely focused electron beam, and the beam's deflection vector due to local electrostatic fields is measured. This allows us to reconstruct an electric field vector map with a resolution defined by the microscope's probe size of 80 pm, which is sufficient to resolve details even within a single unit cell of GaN. However, although the high brightness gun already yields images with excellent S/N, the noise is still problematic for further data processing. Therefore, we use a cross-correlation averaging technique to add identical patches of the crystal's image to further reduce the S/N ratio. This procedure is repeated for every one of the four segments of the DPC detector, and by taking the difference signal between adjacent sectors we obtain a noise-free directional signal which in effect is a projection of the local electric field vector within the specimen on an orthogonal coordinate system defined by the detector. This allows the reconstruction of an electric field vector map of local fields within the GaN unit cell and subsequently the reconstruction of a 2D charge density map from the crystal by using . Fig.1 shows the DF image, giving the atomic locations (left), the reconstructed vector field map (center) and the reconstructed charge density map (right, some atomic positions are indicated by circles). Obviously, the N atoms are much more clearly visible in the charge reconstruction compared to the pure atomic (DF) image. From a detailed evaluation of the experimental charge density image as well as from a DFT calculation one can conclude that there is a charge transfer from Ga towards N which is in agreement with the highly polar covalent type of bonding known to exist in GaN. In Fig. 2, normalized line scans are shown along the line indicated in the inset, showing the DF signal (green, cross-hatched), the charge density signal (blue), the vertical (red, hatched) and horizontal (green) beam deflection as determined by DPC. Clearly, the charge density is peaked at both the Ga and N atomic sites. Please note that the charge density is shifted by a small amount (ca. 9 pm) with respect to the DF signal, which indicates the location of the nucleus. The vertical deflection of the beam is asymmetric around the Ga site which is due to local electrostatic fields. In conclusion we are able to reconstruct charge density distributions within a single unit cell of GaN.

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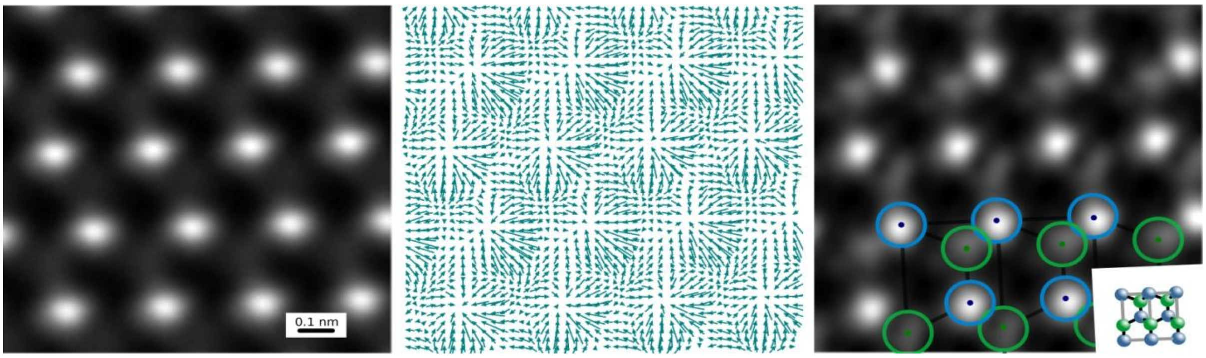

Figure 1. DF image showing atomic locations (Ga atoms. left). Center: reconstructed electric field map of same area as left and (right) reconstructed charge density map with indicators of atomic locations (Ga: blue, N: green).

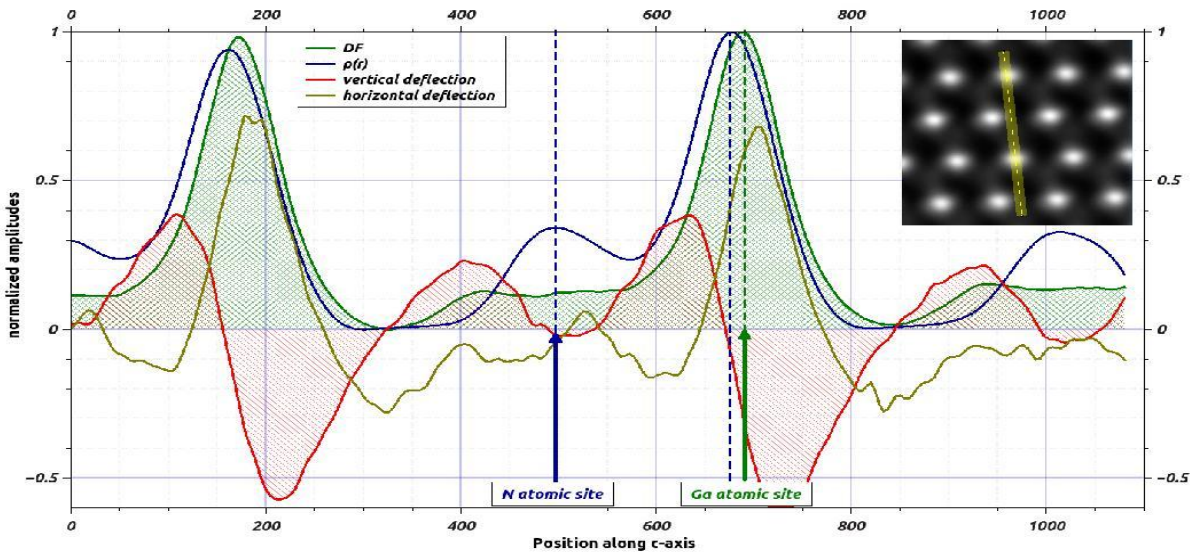

Figure 2. Line scans along the line indicated in the inset image. The DF signal (green) indicates the location of the atomic nuclei, the blue graph shows the reconstructed charge density distribution. Red and yellow curves show the vertical and horizontal beam deflection due to inner electrostatic fields, respectively.