

Simultaneous iDPC and ADF STEM Imaging at the Limit of Contrast and Resolution

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It has recently been reported that the Integrated Differential Phase Contrast STEM (iDPC-STEM) technique [1, 2] enables imaging of both light and heavy atoms together in a thin sample at sub-Å resolution [3]. Furthermore it has been demonstrated that iDPC-STEM obtains phase images of a size and at the same speed as any conventional STEM technique. Therefore, simultaneous ADF-STEM together with iDPC-STEM has become a standard. This way of acquisition provides complementary information on the sample as well as extra flexibility for focusing, stigmation and sample manipulation.

Here we present and further analyze examples of STEM imaging at the limit of resolution and contrast. We use the two above mentioned techniques simultaneously to resolve the atoms at sub-Å scale. As an example, Fig. 1 shows ADF-STEM and iDPC-STEM images for a Wurtzite GaN crystal in $[10\bar{1}1]$ projection as they appear on the screen during acquisition. In this projection gallium and nitrogen dumbbells have a separation of 63 pm. As can be seen in Figure 1a, in the ADF-STEM image Ga atomic columns are visible while Nitrogen columns are absent. On the other hand the iDPC-STEM image displayed in Fig 1.b shows that both Ga-Ga and N-N columns are present and well separated.

Furthermore we study the expected contrast of Ga-Ga and N-N atomic columns and make a quantitative comparison of the ratios of the measured intensities. This comparison shows a very good match with theoretical predictions based on simulations. We use the fact that both ADF- and iDPC-STEM have a very well defined positive definite contrast transfer function (CTF) and we correct for it. The result is shown in Fig. 2. For completeness, a corresponding DPC vector image is also added (Fig. 2c). The color representation indicates the DPC vector direction and the color intensity its modulus.

A quantitative study of the visibility of the N atomic columns between the two techniques reveals that, since with ADF-STEM the square of the electrostatic potential is imaged [2, 4], the resulting signal from atomic columns is roughly proportional to the square of the atomic numbers, Z . The resulting contrast ratio between Ga and N columns is therefore roughly $(31/7)^2 \approx 20$. On the other hand, with iDPC-STEM the electrostatic potential itself is imaged [1, 2] and is roughly linear in Z . Therefore the contrast ratio in this case is $(31/7) \approx 4.5$. This means that a higher dynamic range is needed in ADF-STEM to reveal the N columns together with the Ga columns. The dynamic range of the detector is large enough to accommodate the factor 20 but achieving the necessary SNR is challenging given the weak DF signal.

[1] I. Lazić, E.G.T. Bosch and S. Lazar, *Ultramicroscopy* **160** (2016) 265-280.

[2] I. Lazić, E.G.T. Bosch, *Advances in Imaging and Electron Physics* **199** (2017) 75-184.

[3] E. Yücelen, I. Lazić, E.G.T. Bosch, *Scientific Reports* **8** (2018) 2676.

[4] E.G.T. Bosch, I. Lazić, *Ultramicroscopy* **156** (2015) 59-72.

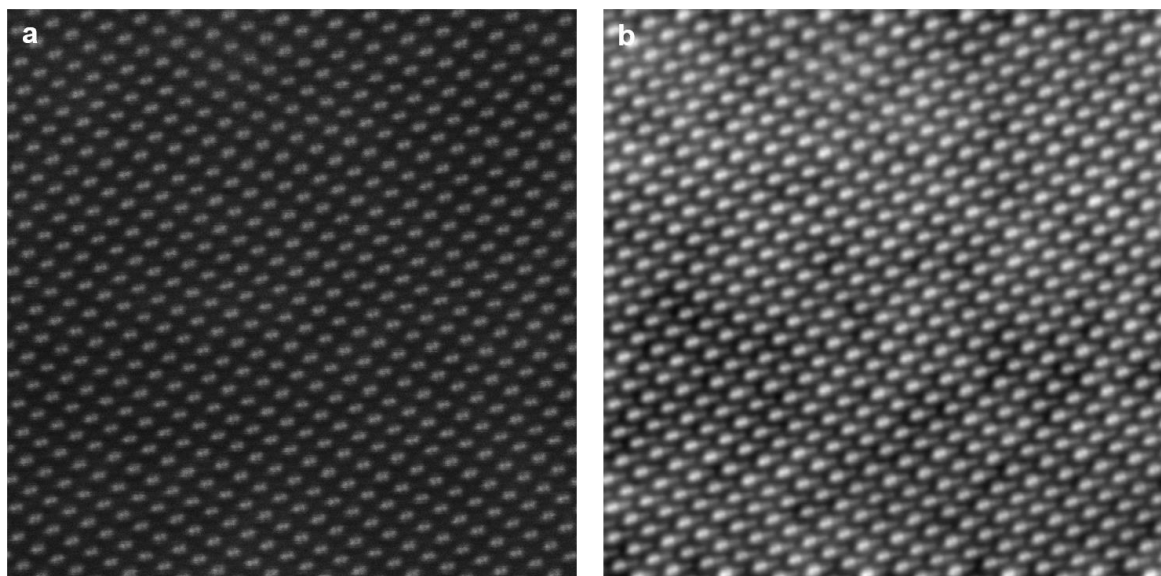


Figure 1. (a) ADF-STEM and (b) iDPC-STEM image of a Wurtzite GaN crystal in $[10\bar{1}1]$ projection. Images acquired at 300 kV with convergence semi-angle of 30 mrad. The field of view (FOV) is 6 nm.

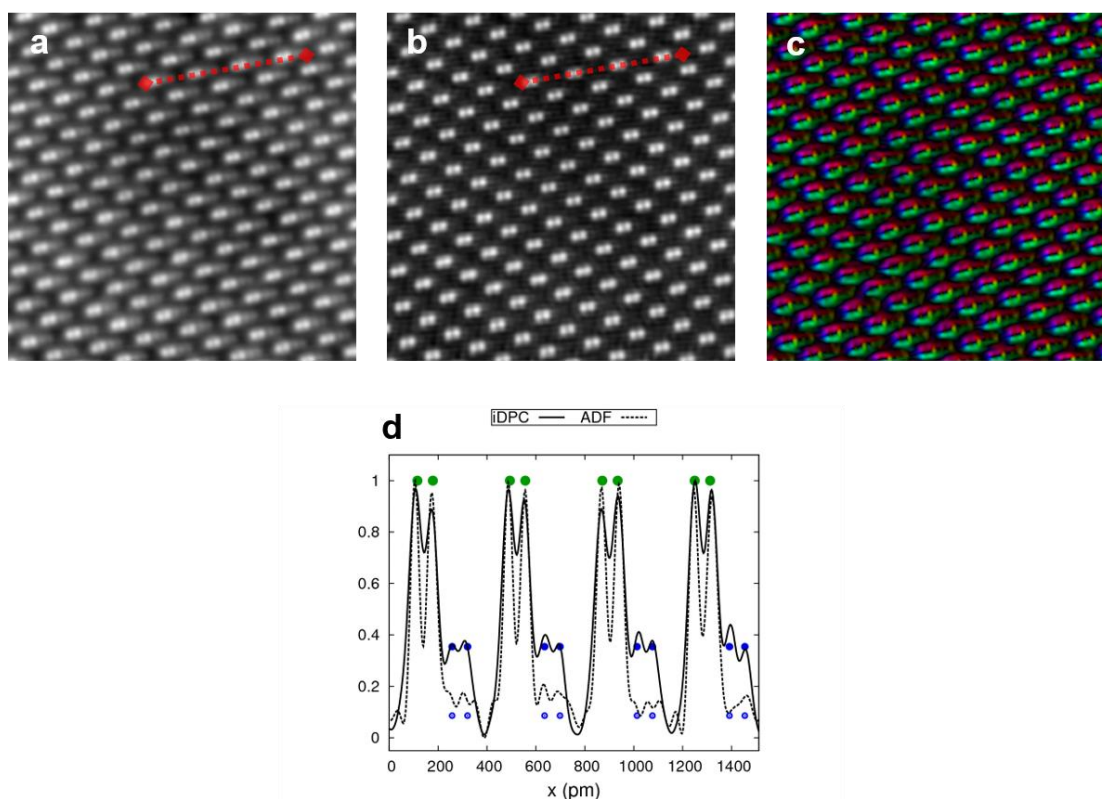


Figure 2. (a) iDPC-STEM (b) ADF-STEM and (c) DPC vector image of GaN in $[10\bar{1}1]$ orientation. Images are CTF corrected. The FOV is 3 nm. (d) Normalized intensity profile plots of iDPC- (solid line) and ADF-STEM (dashed line) along the indicated red dashed lines. Green dots: position and expected intensity of the Ga columns. Solid blue dots: expected intensity of the N columns in the iDPC signal. Open blue dots: expected N column intensity of the ADF-STEM image.