

## **Spatial resolution of 40.5 pm by using scanning transmission electron microscopy with fifth-order aberration corrector**

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Sub-Angstrom spatial resolution has become routinely available by using transmission electron microscopy (TEM) and scanning TEM (STEM) with aberration correction. Aberration correctors expanded convergence/acceptance angles in S/TEM and improved the diffraction limit. To date, the best spatial resolution is 45 pm using Si [114] in STEM [1] and 44 pm using GaN [411] in TEM [2]. Such high-resolution imaging can be applied to the determination of atomic positions and also to the detection of an electric-field distribution of each atom. For precise measurement of the interatomic field observations, the spatial resolution is still insufficient and further improvement of the resolution is strongly required.

To improve the spatial resolution in STEM, the diffraction limit has been improved by developing geometrical aberration correctors. However, the semi-angle was limited to approximately 30 mrad at middle accelerating voltages. The diffraction limit is still dominant limiting factor for the spatial resolution and should be improved. In the present study, we have developed 300-kV STEM equipped with a delta-type corrector. The delta-type corrector can correct geometrical aberrations up to fifth order including six-fold astigmatism, which is the dominant aberration in the standard double-hexapole type corrector. The delta-type corrector has originally been developed for a low accelerating voltage microscope [3]. We here newly designed a delta-type corrector for 300-kV microscope and installed to GRAND ARM™ (JEM-ARM300F). After an optimum alignment of whole lens system, sixth-order three-lobe aberration, which is a dominant aberration in the delta-type corrector [4], was measured to be 1.8 mm. A contrast flat area in a Ronchigram expanded to 70 mrad at 300 kV, which is almost twice the conventional aberration-corrected angle.

By using this microscope, we demonstrated the resolution improvement using the specimen of GaN [212]. The observation was performed with several convergence angles to investigate the effect of the diffraction limit. Annular dark-field images of GaN [212] with convergence semi-angles of 20 and 30 mrad showed only broad peaks with a separation of 243 pm. With the larger convergence angle of 40 mrad, we are able to resolve a dumbbell structure of Ga-Ga atomic columns with a spacing of 40.5 pm (Fig.1a,b). In the intensity profile, the dip between two dumbbell peaks is estimated to be 13% (Fig.1c). This result indicates that the improvement of the diffraction limit is still essential for the enhancement of the resolution. We experimentally demonstrated that the spatial resolution of 40.5 pm can be achieved by using the delta-type corrector at 300 kV [5]. This supremely high spatial resolution imaging could be advantageous to observe complicated atomic structures and inter-atomic fields.

[1] Sawada H, et al., (2015) *Microscopy* 64: 213 - 217.

[2] Akashi T, et al., (2015) *Appl. Phys. Lett.* 106: 074101.

[3] Sawada H, et al., (2009) *J. Electron. Microsc.* 58: 341 - 347.

[4] Morishita S, et al., (2018) *Microscopy*.

[5] Morishita S, et al., (2017) *Microscopy* 67: 46 - 50.

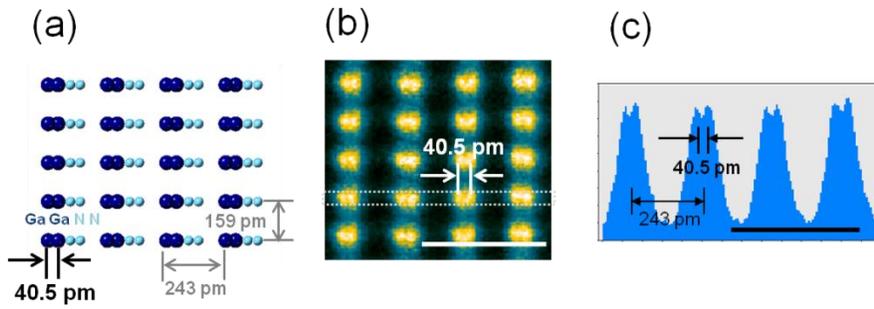


Figure 1. (a) Atomic model of GaN along the [212] direction. (b) ADF STEM image of GaN [212] with a convergence semi-angle of 40 mrad at 300 kV. The image was averaged using 10 images taken with a dwell time of 38  $\mu$ s per pixel. (c) Intensity profile along the dotted line in (b). Scale bars in (b) and (c) indicate 0.5 nm.