Supplementary Information (SI) for Nanoscale.

Supporting Information

Thickness-Dependent Polarization Modulation at AlN Interlayers in GaN Heterostructures Revealed by Atomic-Scale 4D-STEM

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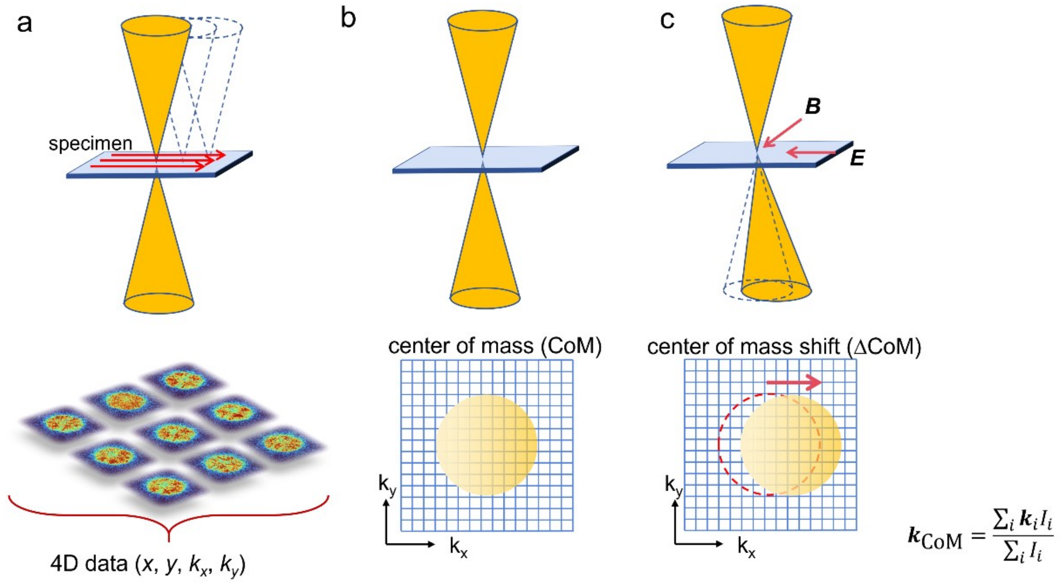

Figure S1. Schematic diagrams of the principles of 4D-STEM and CoM. (a) Each probe position on the sample corresponds to a two-dimensional CBED collected by the pixelated detector. By scanning the sample, a set of 4D data containing information in real space and reciprocal space can be obtained. (b) The CoM vector is calculated by performing a weighted summation of the direct beam spot and then divide by the total intensity. (c) The schematic diagram of CoM shift. When an electric or magnetic field exists within the sample, the electron beam transmitted through the sample will be deflected. By calculating the difference between the CoM vector of the deflected beam and the average CoM vector, the CoM shift can be obtained.

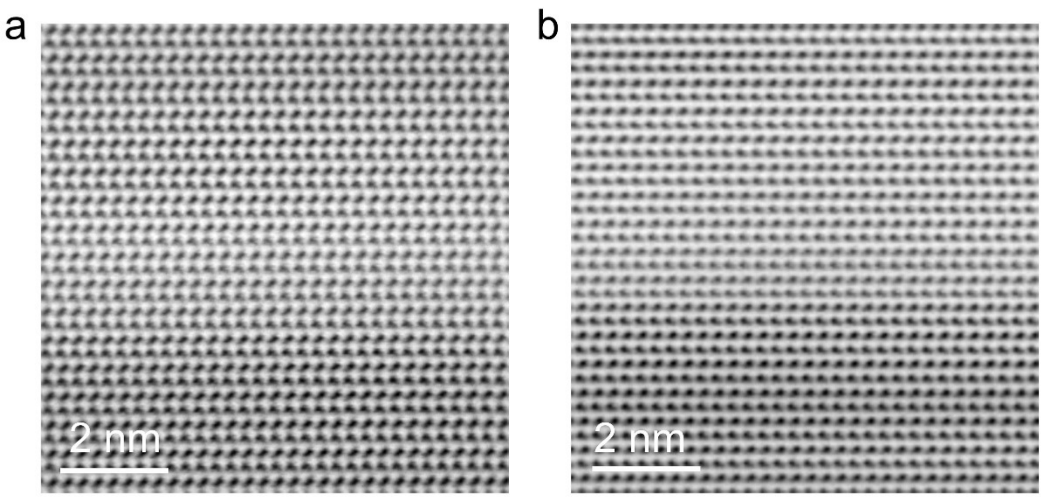

Figure S2. ABF-STEM images via conventional STEM of the samples with 1 nm (a) and 0.5 nm (b) AlN interlayers.

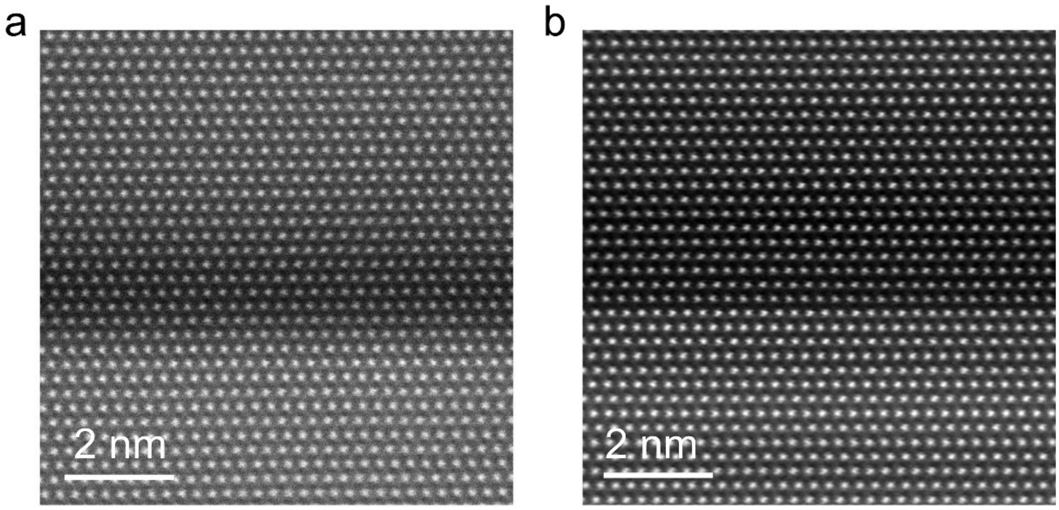

Figure S3. HAADF-STEM images of the samples with 1 nm (a) and 0.5 nm (b) AlN interlayers viewed from $[11\bar{2}0]$.

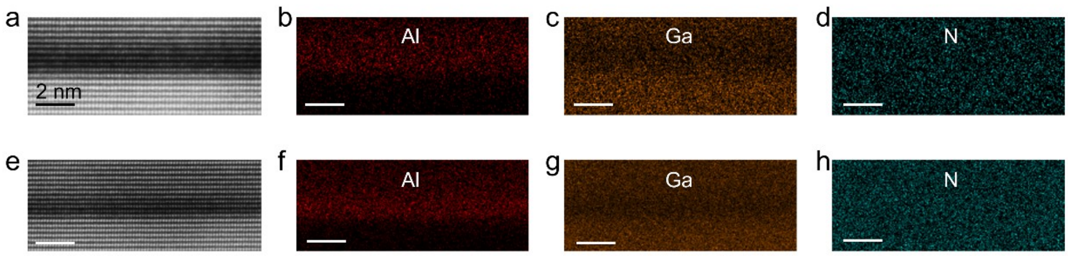

Figure S4. EDS mapping of heterojunction with 1nm (top panel) and 0.5 nm (bottom panel) AlN interlayers. (a, e) HAADF-images of the top three-layer interfaces in the epitaxial structure with 1 nm (a) and 0.5 nm (e) AlN interlayers, respectively. (b-d, f-h) elemental distribution mappings of Al, Ga and N of (a) and (e), respectively. The scale bar is 2 nm for all the images.

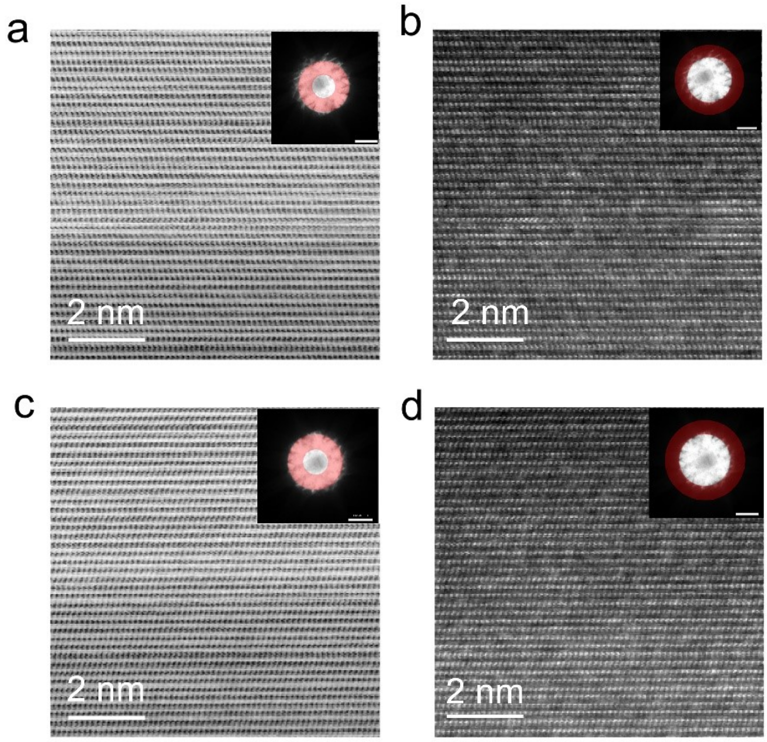

Figure S5. Reconstruct images from 4D datacubes with 1nm (top panel) and 0.5 nm (bottom panel) AlN interlayers. (a, c). Reconstruct ABF-STEM images from 4D datacubes by masking and integrating to the corresponding detector angles (Mask: 65 ~ 90 mrad) on each Ronchigram. (b, d) Reconstruct LAADF-STEM images from 4D datacubes by masking and integrating to the corresponding detector angles (Mask: 30 ~ 65 mrad) on each Ronchigram. The scale bar of all the inset images is 1 nm^{-1}.

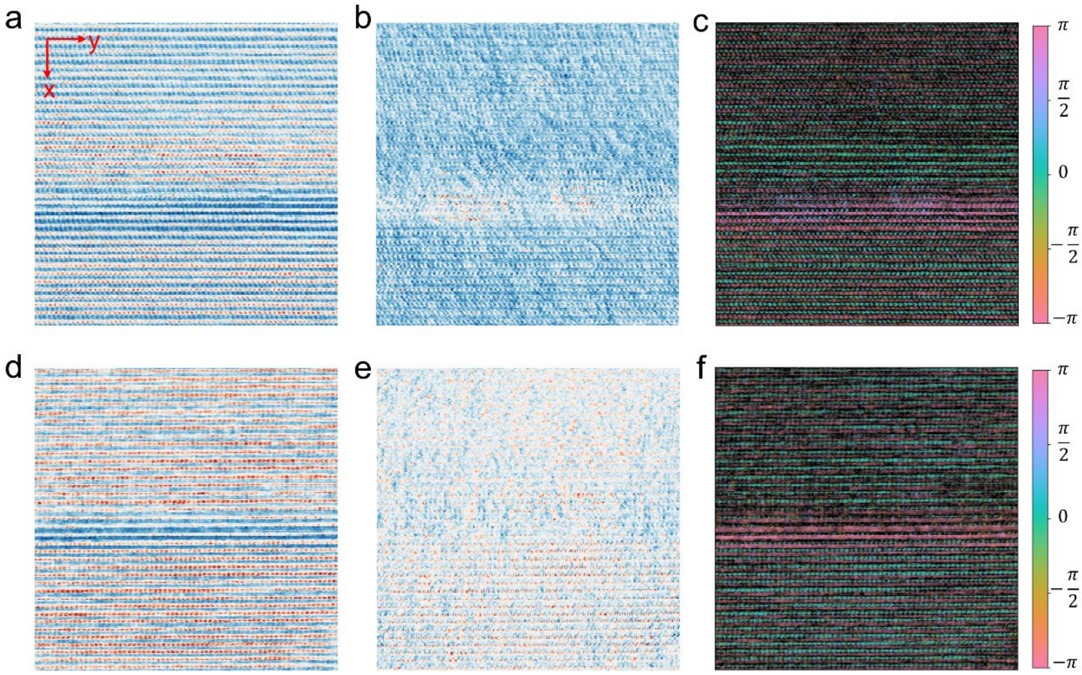

Figure S6. Experimental 4D-STEM CoM shift measurements heterojunction with 1nm (top panel) and 0.5 nm (bottom) panel AlN interlayers. (a, d) CoM shift vectors alone x directions at each pixel positions. (b, e) CoM shift vectors alone y directions at each pixel positions. (c, f) The directions of CoM shift vectors at each pixel positions.

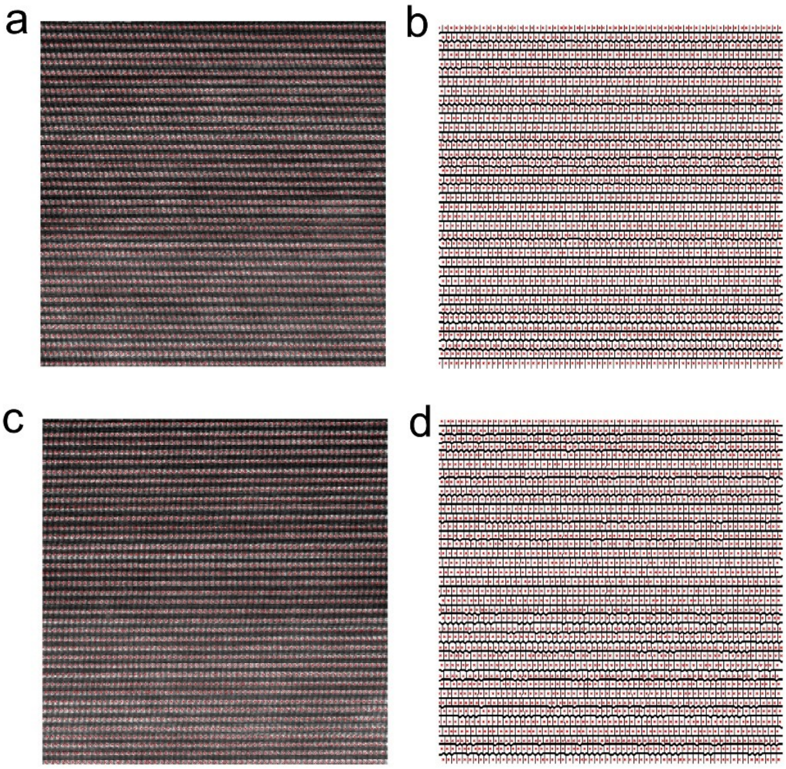

Figure S7. Voronoi diagram division of heterojunction with 1nm (top panel) and 0.5 nm (bottom) panel AlN interlayers. (a, c) Atomic positions identification (marked by red crosses) on the basis of Figure 3a and e, respectively. (b, d) Voronoi diagrams, where each cell in enclosed by a black frame, and the red crosses indicate the atomic positions.

Calculation of the sample thickness

Low-loss EELS spectra contain zero-loss (I_0) and plasmon-loss (I_p) peaks, as illustrated in the accompanying images. The sample thickness Δz can be derived using the following equation[1, 2]:

$$\Delta z = \lambda \ln \frac{I_t}{I_0} (S1)$$

where λ denotes the mean free path for inelastic electron scattering, and I_t is the total integral of the EELS spectrum. The mean free path λ is calculated via the equation below:

$$\lambda(nm) = \frac{106F\left(\frac{E_0}{E_m}\right)}{\ln\left(\frac{2E_0\beta}{E_m}\right)}, F = \frac{1 + E_0/1022}{\left(1 + \frac{E_0}{511}\right)^2}, E_m = 7.6Z_{eff}^{0.36} (S2)$$

Here, E_0 is the incident electron energy in keV, F is a relativistic factor, β is the collection semi-angle in mrad, and Z_{eff} is the effective atomic number. Under the present experimental conditions,

E_0 = 200 keV, F = 0.618, and β = 16.6 mrad. The effective atomic number Z_{eff} is determined by:

$$Z_{eff} = \frac{\sum_i f_i Z_i^{1.3}}{\sum_i f_i Z_i^{0.3}} \quad (S3)$$

where f_i is the atomic fraction of each element which has atomic number Z_i. Z_{eff} is calculated to be approximately 18.3. In the AlGaN/AlN/GaN heterostructure system, the Z_{eff} of GaN was adopted as a substitute for that of the entire system, yielding a mean free path of ~89.5 nm for GaN.

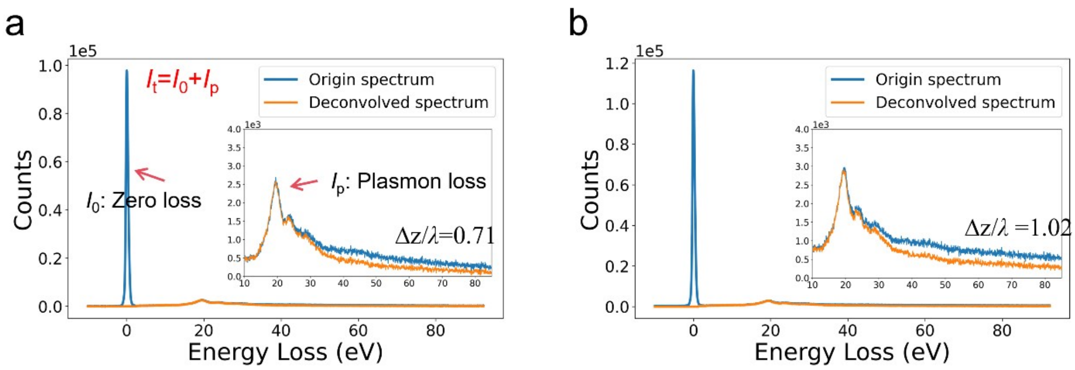

Figure S8. Low loss spectrums with 1 nm (a) and 0.5 nm (b) AlN interlayers, respectively. The blue spectral line represents the original data, while the yellow spectral line corresponds to the result after deconvolution to remove plural scattering. The inset are the enlarged spectrums of 10 ~ 85 eV.

References

(1) Malis, T.; Cheng, S.; Egerton, R. EELS log-ratio technique for specimen-thickness measurement in the TEM. *Journal of electron microscopy technique* **1988**, *8* (2), 193-200.

(2) Heo, Y.-U. Comparative study on the specimen thickness measurement using EELS and CBED methods. *Applied Microscopy* **2020**, *50* (1), 8.