Unravelling the polarity of InN Quantum Dots using a modified approach of negative-spherical-aberration imaging

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Electronic Supplementary Information

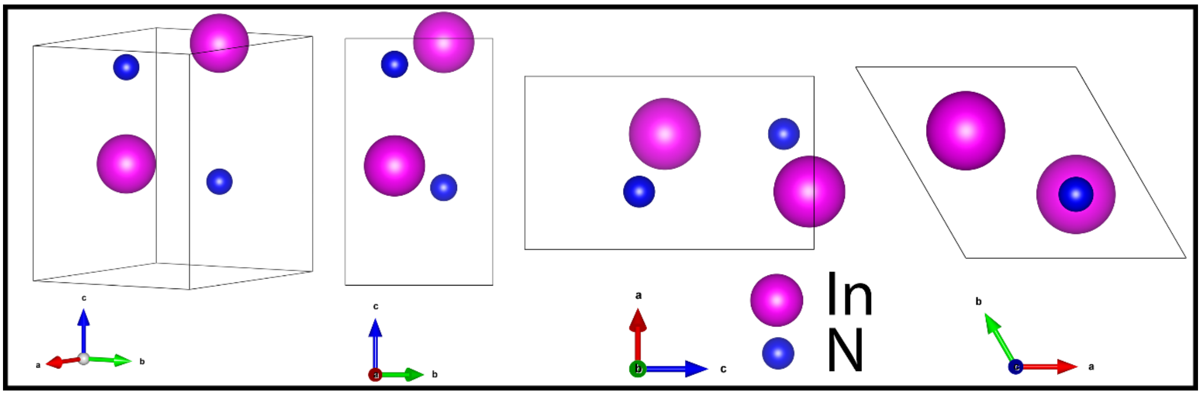

Fig. S1: Relaxed unit cell of Wurtzite InN structure as viewed in VESTA in different orientations.

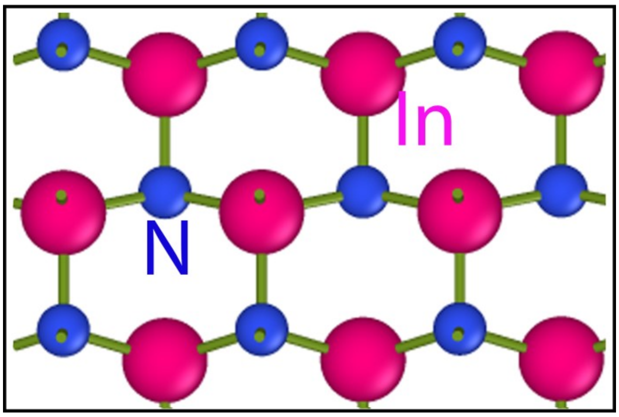

Fig. S2: Model of N polar InN structure. Blue circles represent N atoms and pink circles represent In atoms.

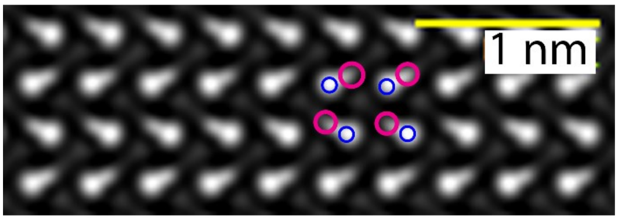

Fig. S3: Simulated HRTEM image along [11$\bar{2}$0] of In polar InN slab with thickness of 8 nm along the viewing direction with same experimental parameters and at 6.7 defocus similar like the N polar one shown in left inset of Fig. 2. Blue circles represent N atoms and pink circles represent In atoms.

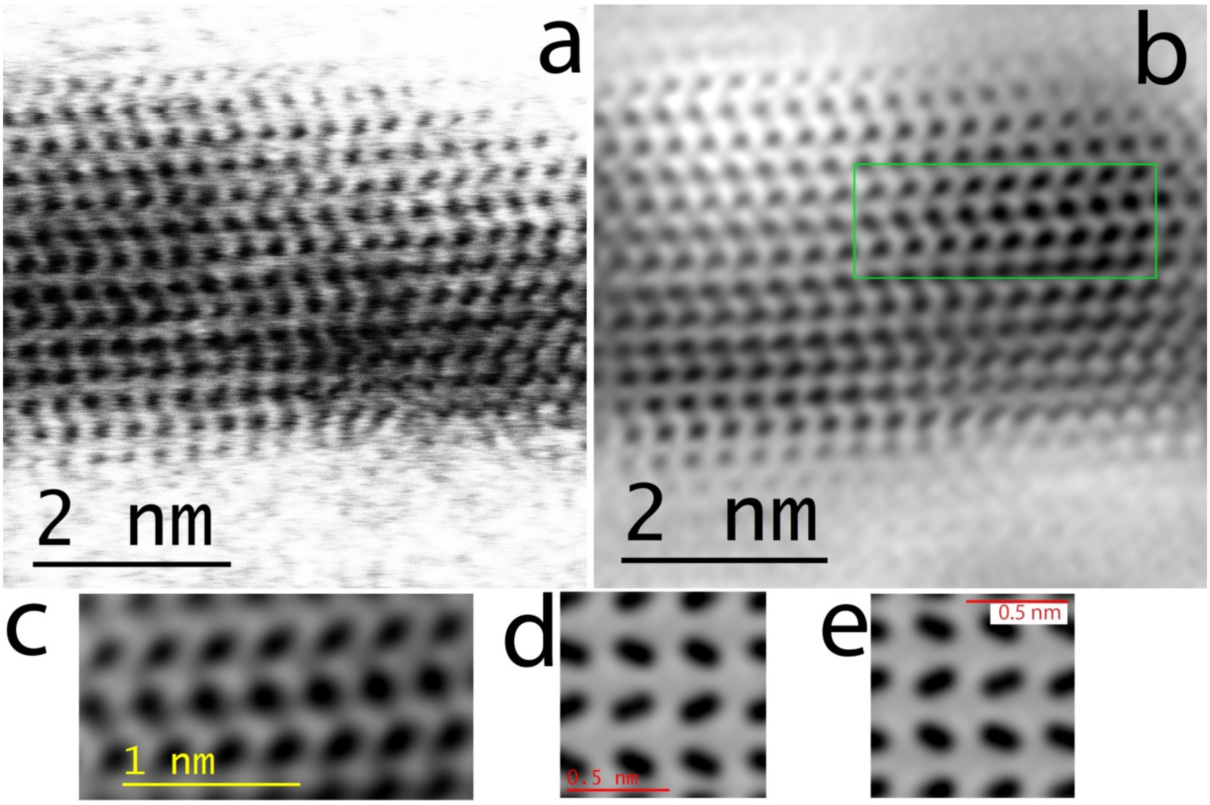

Fig. S4. (a) Experimental STEM image recorded on an annular detector with a collection semiangle of 12.7 to 24 mrad of an InN quantum dot along [11$\bar{2}$0] zone axis. (b) Same image after background subtraction using algorithm[3], (c) zoomed version of the region marked in green colour in (b); simulated STEM images of InN block of thickness 8 nm along [11$\bar{2}$0] with same parameters as experiment using effective source size 0.9 Å with (d) N polarity and (e) In polarity.

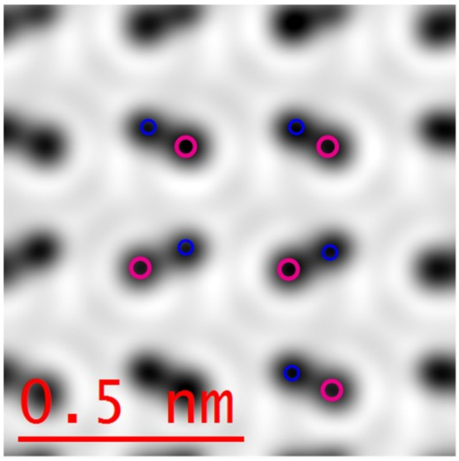

Fig. S5: Simulated STEM image on an annular detector with collection semiangle 12.7 to 24 mrad of N polar InN block of thickness 8 nm along $[11\bar{2}0]$ with effective source size 0.6 Å. This simulation was done with 200 kV accelerating voltage and 0.6 µm Cs of condenser lens. Blue circles represent N atoms and pink circles represent In atoms.

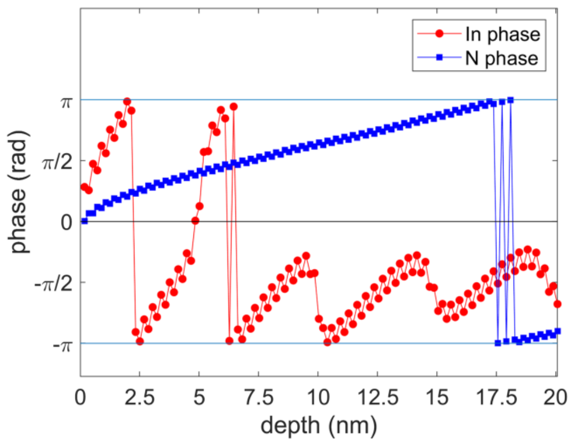

Fig. S6. Peak phase values of In and N columns from simulated exit face wave with varying thickness along $[11\bar{2}0]$ of a InN slab.

Indium (atomic no. 49) is much heavier atom than Nitrogen (atomic no. 7). We have simulated InN slab of varying thicknesses along $[11\bar{2}0]$, propagated plane wave through the specimen and determined exit waves (above the objective lens of TEM) at different thicknesses. Then we have plotted peak phase value of In and N columns atomic positions w.r.t specimen thickness which is shown in Fig. S6. In the above mentioned figure depth represents the specimen thickness along the viewing direction $[11\bar{2}0]$.

Fig. S6 reveals that with increasing thickness, phase value of N increases linearly while In has phase inversion very often. Therefore, without knowing the exact thickness along the viewing direction for InN it is very difficult to confidently unwrap the reconstructed phase.

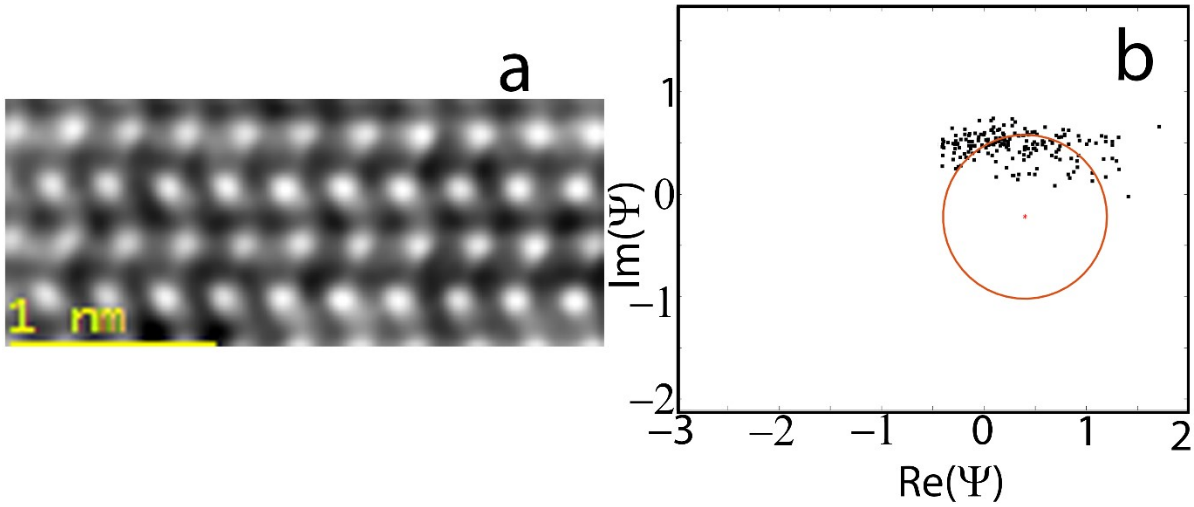

Fig. S7. Reconstructed (a) phase shown in Fig. 1c from the region marked with green coloured box in Fig. 2. (b) Argand-Gauss diagram of the experimental exit wave of first QD using bright peak positions.

Comparing Fig. S7a with Fig. 2 the position of In atomic columns were determined which show In has negative phase value. 2π value was added to these In positions that brings these peak phase values higher than that of N peaks which is expected from their respective atomic potentials. As an example, the reconstructed phase value In feature in first row 3 column (from the left) of Fig. S7a is −0.2 rad which after adding 2π becomes higher than peak phase value (0.49 rad) of N feature of the same dumbbell.

The determined complex constant offset for the reconstructed complex wave from Fig. S7 c is 0.4−i0.22.