Supplementary Information

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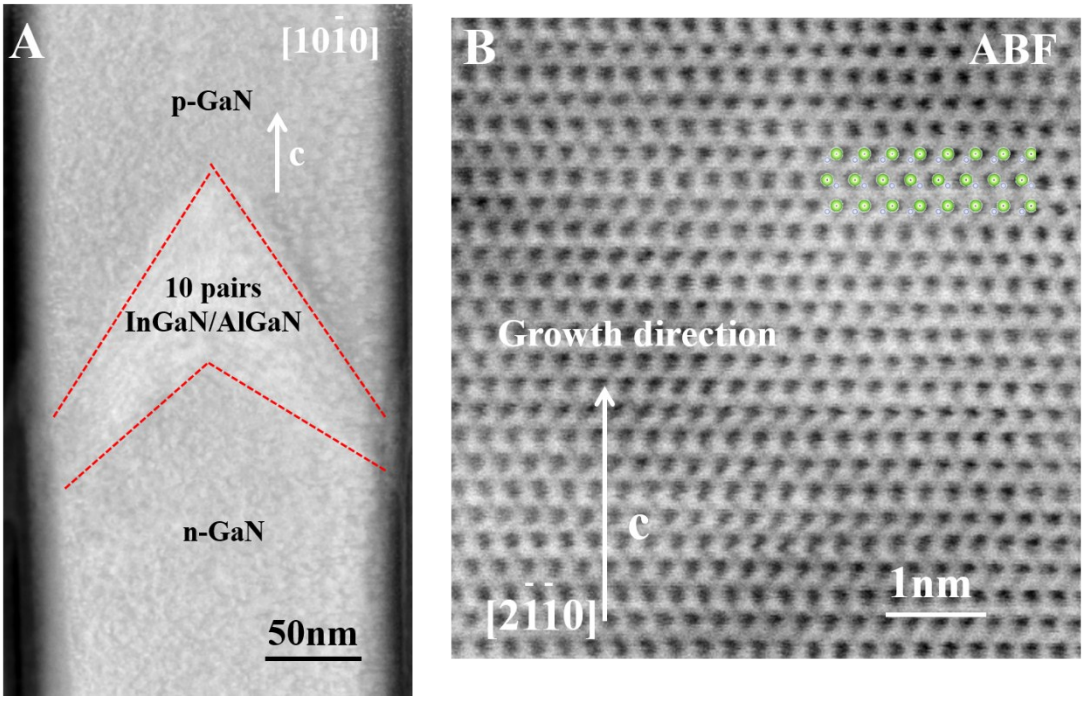

Fig. S1 STEM image of the QDs. (A) HAADF-STEM images from $[10\bar{1}0]$ zone axis. No contrast from individual QDs can be observed. (B) Annular bright field (ABF) image acquired from GaN nanorod showing the Ga-face nature. The atomic model of GaN from $[2\bar{1}\bar{1}0]$ zone axis is overlaid.

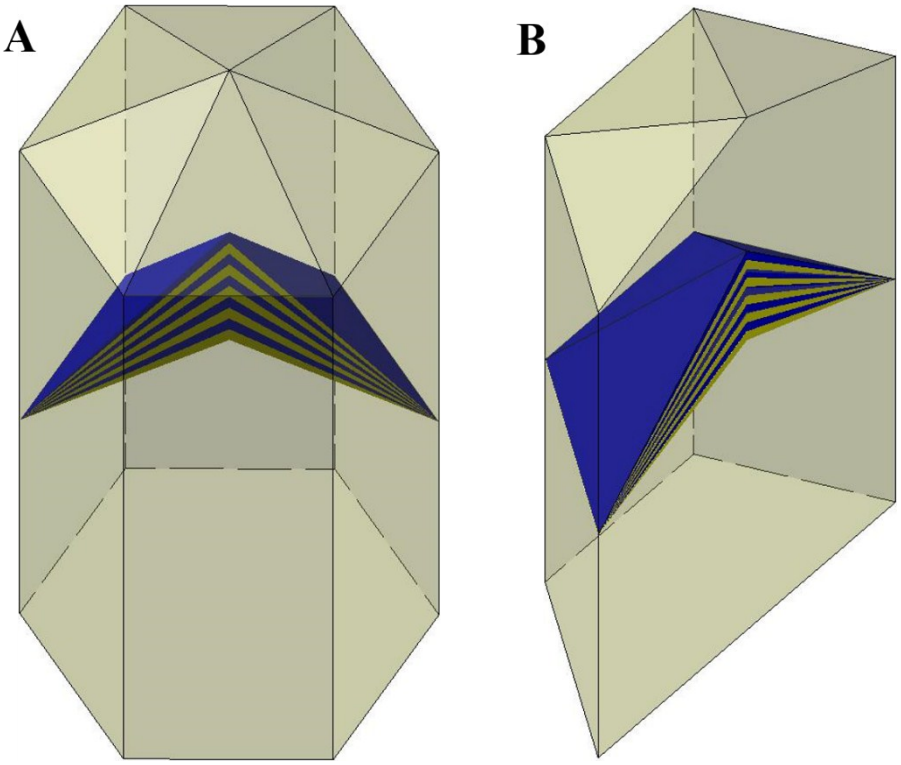

Fig. S2 Three-dimensional view of the boomerang shape QD. The starting planes for the QDs (the bottom quantum dot of the series) are $\{10\bar{1}2\}$, while the ending planes at the top are $\{10\bar{1}1\}$. The very top surfaces of the GaN nanowires (before the growth of the QDs and at the very top of the wire) are composed by $\{10\bar{1}2\}$ planes.

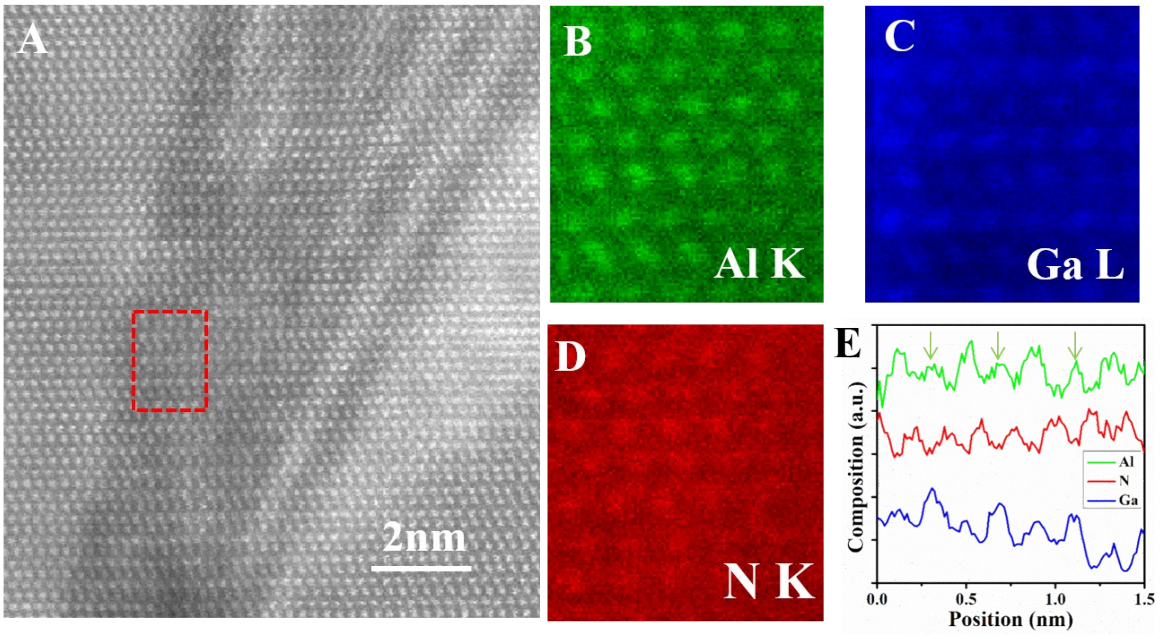

Fig. S3 Composition mapping by EELS. (A) The ADF signal collected concurrently with the EELS mapping. (B-D) The EELS mapping results of Al K edge, Ga L edge, N K edge for the red rectangle area in (A). (E) Relative composition in the same area. The green arrows mark the Ga-rich layers. The modulated concentration of Al and Ga can be clearly observed.

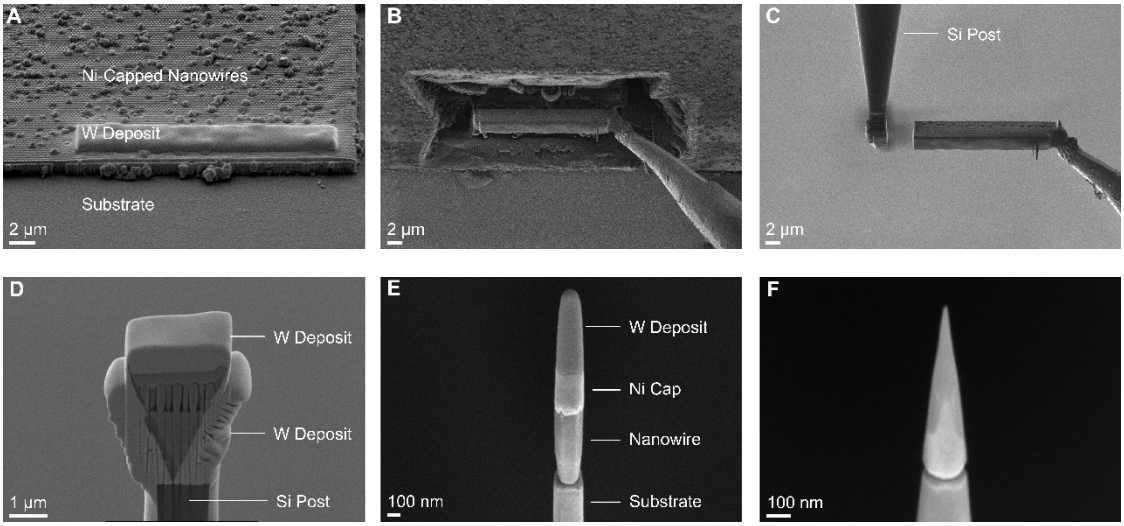

Fig. S4 FIB sample preparation of APT specimens. Patches of nanowires were first protected by sputter-coating with ~200 nm of Ni. An area approximately 20 x 3 μm was selected and coated with a W deposit (A). This region was cut out and extracted using a micro-manipulator (B). Sections of the lift-out were attached to the tips of pre-fabricated Si posts using W deposition, then cut off from the liftout (C) making a total of 6 needles with a patch of nanowires on the top (D). Each needle was trimmed down so that only a single nanowire was left on each post (E). The remaining nanowire was sharpened using a defocused, low-energy beam until no W or Ni remains on the tip (F).

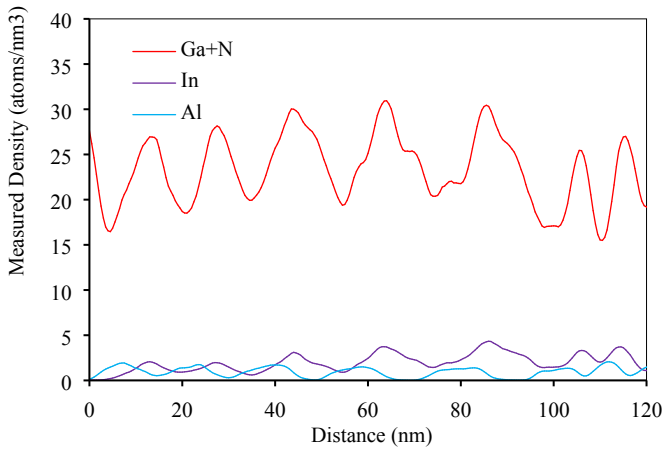

Fig. S5 The measured sample density by APT. Compositions as measured by APT from the QDs are complicated by trajectory aberrations, which can cause overlap of ions between layers (e.g. Al in the InGaN layer). To improve measurement accuracy, In and Al atoms are segregated based on individual layers, so as not to alter calculated compositions (i.e. Al is excluded in the InGaN layer, and vice versa). The data are sampled from a sub-volume ROI with 10 x 10 nm^2 cross-section, extending down within the center of a "facet". Layers are delineated by local minima in the In or Al atom density profile, and by inflection points in the GaN density profile (see above right, where the top QD is at x = 0).

Table S1 Composition results, using the methodology described above, for each of the InGaN and AlGaN QDs. The Layer No. 1 is the topmost layer.

Layer	InGaN Layer (at.%)			AlGaN Layer (at.%)		
No.	**In**	**Ga**	**N**	**Al**	**Ga**	**N**
1	8.3	39.5	52.1	8.1	41.5	50.4
2	9.8	39.8	50.4	8.7	38.3	53.0
3	9.1	39.0	51.8	12.9	38.2	48.9
4	13.1	36.4	50.5	8.6	39.9	51.2
5	16.9	33.6	49.5	6.6	39.0	54.3
6	19.2	33.1	47.7	5.5	39.2	55.3
7	19.5	35.1	45.4	10.9	33.7	55.4
8	15.2	34.2	50.5	13.2	35.7	51.1