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Supplementary information

Highly elongated vertical GaN nanorod arrays on Si substrates with AlN seed layer by pulsed-mode metalorganic vapor deposition

Si-Young Bae[1, *], Byung Oh Jung[1], Kaddour Lekhal [1], Sang Yun Kim[2,3], Jeong Yong Lee[2,3], Dong-Seon Lee[4], Manato Deki[1], Yoshio Honda[1], and Hiroshi Amano[1,5]

[1]Department of Electrical Engineering and Computer Science, Nagoya University, Nagoya, Aichi 464-8603, Japan

[2]Center for Nanomaterials and Chemical Reactions, Institute for Basic Science (IBS), Daejeon 305-701, South Korea

[3]Department of Materials Science and Engineering, Korea Advanced Institute of Science and Technology (KAIST), Daejeon 305-701, South Korea

[4]School of Information and Communications, Gwangju Institute of Science and Technology (GIST), Gwangju 500-712, Korea

[5]Akasaki Research Center (ARC), Nagoya University, Nagoya, Aichi 464-8603, Japan

[*] Corresponding author e-mail: siyoubae@gmail.com (S.-Y. Bae)

1. **Pulsed-mode sequence**

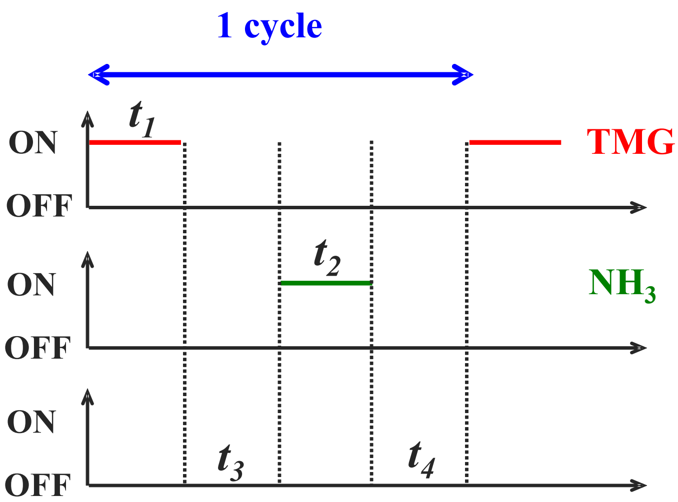

Figure S1. Schematic of the pulsed-mode sequence showing the TMG injection time (t_1), NH_3 injection time (t_2), post-TMG interruption time (t_3), and post-NH_3 interruption time (t_4).

2. Length distribution of grown nanostructures

Figure S2 shows the height and diameter distributions of the grown nanostructures illustrated in Fig. 1. From its definition, PMGR can be increased by decreasing the TMG injection time (t_1) or increasing the NH_3 injection time (t_2). Height became much smaller as PMGR increased from the optimized condition (PMGR = 3). On the other hand, as PMGR decreased from PMGR of 3, broader distribution of the height and diameter were observed by error bars in length, thereby resulting in the disordered structures in both the vertical and lateral directions. From the observation of both cases, it was found that we could grow high-aspect-ratio GaN nanorods when PMGR was ~3.

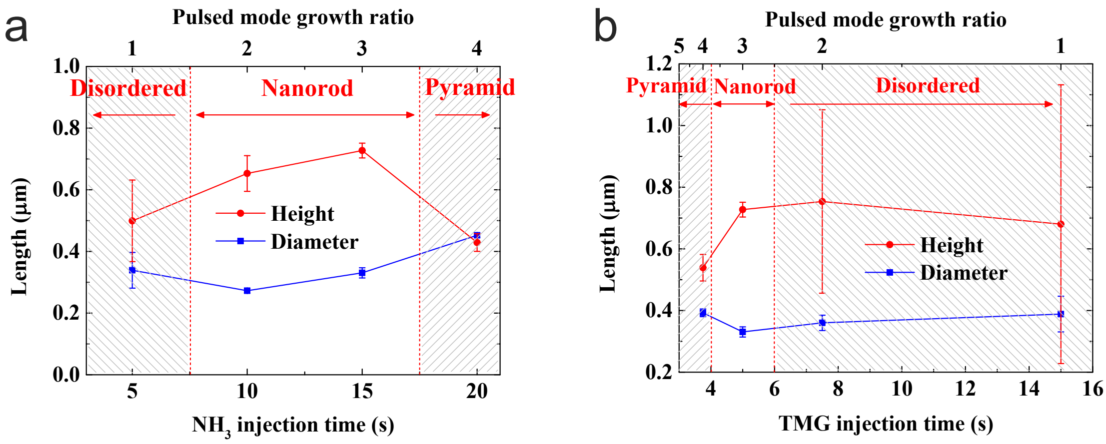

Figure S2. Height and diameter distributions of grown GaN nanostructures as a function of (a) NH_3 injection time and (b) TMG injection time. Note that hexagonal pyramid structures were typically observed for a high PMGR (≥ 4), while disordered structures were found for a low PMGR (≤ 1).

3. Material properties of AlN/Si template

Figure S3 shows the various material properties of the AlN/Si template. The AlN layer had a slightly rough morphology, as can be seen from the SEM image in Fig. S3a, and the RMS roughness was 1.84 nm (Fig. S3b). By measuring the rocking curve of the XRD omega scan, the FWHM was determined to be 1.72°. Owing to its low thickness of ~50 nm, the AlN layer was of low quality. On the other hand, its in-plane and out-of-plane preferential orientations retained their directionality, as shown in the EBSD images in Fig. S3d.

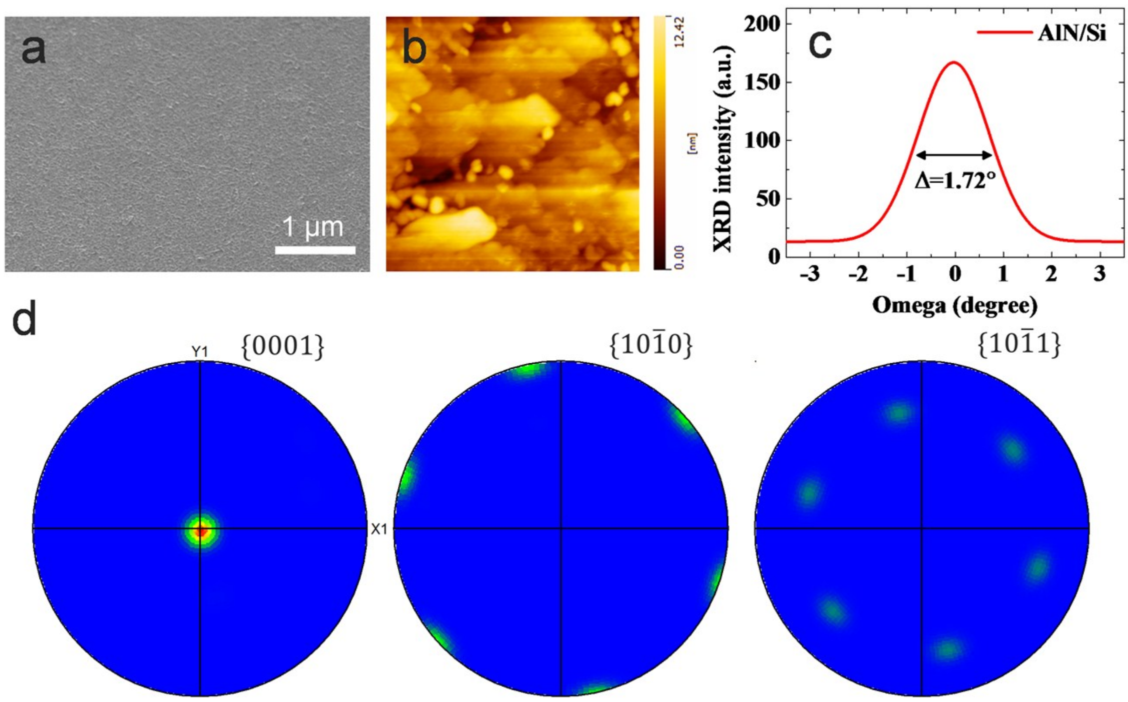

Figure S3. Material properties of AlN on Si used in this study evaluated from (a) SEM image, (b) AFM image, (c) XRD rocking curve, (d) EBSD pole figures in the {0001}, {10$\bar{1}$0}, and {10$\bar{1}$1} planes.

4. Parameter determination for the kinetic Wulff plot

Figure S4 depicts the method used to determine each growth rate in this study. From the cross-sectional view in Fig. S4a, we determined the growth rates in three main directions, i.e., $[0001]$, $[10\bar{1}1]$, and $[10\bar{1}0]$. Here, we used linear interpolation (dotted line) along the $\{10\bar{1}1\}$ semipolar surface and found the point of the intersection with normal line along the $[10\bar{1}1]$ direction. Hence, we assume that the vertical length along the $[0001]$ direction is *directly* related to the growth rate of the $\{10\bar{1}1\}$ planes. To reduce the error in the lateral growth rate, top-view SEM images were also compared, as shown in Fig. S4b. Then, the lateral growth rates along the $[11\bar{2}0]$ and $[10\bar{1}0]$ directions due to vertical growth were measured.

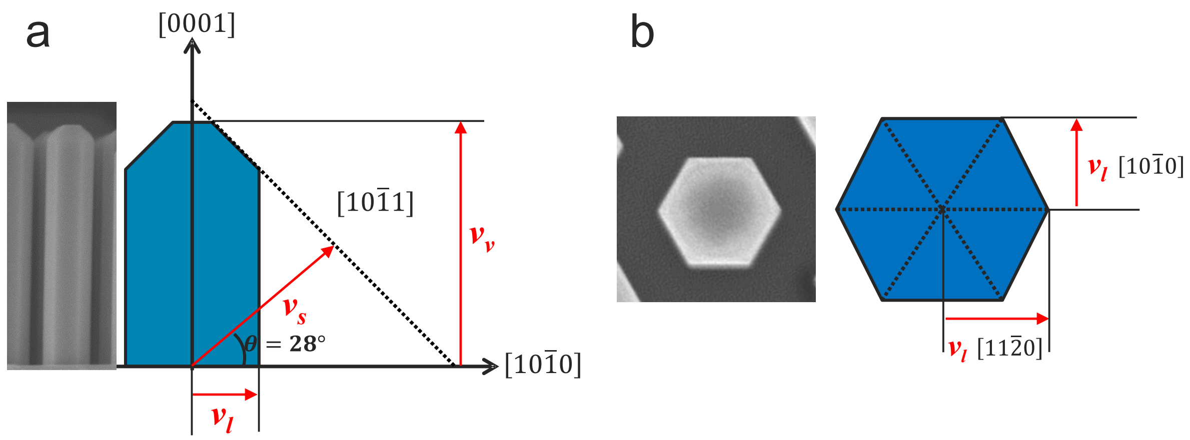

Figure S4. SEM image (left) and schematic diagram (right) used to determine growth rate of each equilibrium surface using (a) cross-sectional view and (b) top view. Depending on the view, the growth rates in several directions could be obtained such as $[0001]$, $[10\bar{1}1]$, $[1010]$, and $[11\bar{2}0]$.