ELECTRONIC SUPPLEMENTARY INFORMATION

Three-dimensional epitaxy of single crystalline semiconductors by polarity-selective multistage growth

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Fig. S1 (a) SEM images showing ZnO hexaplates depending on the NH₄OH concentration (C_{AH}) in the precursor solutions. (b)–(d) SEM images of diverse types of ZnO crystal patterns depending on the point seed arrays. (b) Linear arrays of coalescent ZnO hexaplates grown with point seeds (top) aligned along the [110] (middle) and [100] (bottom) directions. (c) Pac-man like ZnO crystals. (d) Macroscopic ZnO plate (bottom) consisted of ZnO hexaplates grown out from point seed arrays (top). All scale bars are 1 μ m.



Fig. S2 Schematic illustrating the hydrothermal process for multistage growth of 3D structures.



Fig. S3 EBSD pole figures showing the orientation distribution of the coalescent ZnO hexaplate shown in Figure 4b. (a) C-axis plot showing that the basal {001} planes of the ZnO hexaplate are oriented within $\pm 3^{\circ}$ with respect to the substrate surface. (b) The {110} planes show six dominant poles separated by 60°, indicating a six-fold in-plane symmetry.



Fig. S4 X-ray rocking curves of ZnO epitaxial seed (red) layer and ZnO hexaplates (blue). The narrow full width at half maximum value of ZnO hexaplate array (0.5) illustrates that ZnO crystals are epitaxially grown on the ZnO seed layer.



Fig. S5 (a) low-magnification TEM images of 3D tubular structured GaN crystal that was made after etching the core ZnO hexaplate. (b, c) A fragment of GaN crystal and selective area electrondiffraction pattern showing the single crystal characteristics. (d)–(f) The high-magnification TEM images taken from regions I (d), II (e), and III (f) in the image of (b), with the corresponding FFT images (insets).