

Supporting Information for

Synthesis route and growth mechanism of aligned GaN

nanobelts

Chenlong Chen, Mitch M. C. Chou,* Tao Yan, Huichun Huang, Cheng-Ying James Lu, and Chienhui Chen

NSC Taiwan Consortium of Emergent Crystalline Materials, Department of Materials and Optoelectronic Science, National Sun Yat-Sen University, Kaohsiung 80424, Taiwan

Corresponding Author: mitch@faculty.nsysu.edu.tw

Part 1. Experimental Section

Synthesis of the crystallographic aligned GaN nanobelts on (100) γ -LiAlO₂ substrate:

High-quality 2-inch γ -LiAlO₂ single crystals were grown using the Czochralski pulling technique. The (100) γ -LiAlO₂ substrate was polished with root-mean-square roughness of 0.3.2~0.48 nm, subsequently washed in ultrasonic bath of acetone and ethyl alcohol and dried in nitrogen flow. A 5~10 nm thin layer of Au was deposited on the substrate by a Pelco SC-6 sputter coater. A five-temperature-zone horizontal tube furnace CVD system was designed for the growth of nonpolar GaN nanobelt arrays on (100) γ -LiAlO₂ substrate. Briefly, the furnace consists of one fused-quartz outer tube (230 cm in length, 15 cm in outer diameter, and 0.3 cm in wall thickness), two fused-quartz inner tubes (120 cm in length, 5 cm in outer diameter, and 0.25 cm in wall thickness), and one designed fused-quartz utensil for flow merged. The temperature, pressure, atmosphere, and synthesis time were automatic controlled. High-purity metallic Ga (99.9999%) and gaseous NH₃ (99.9995%) were used as Ga

and N source, respectively. Nitrogen was used as a carrier gas. Gallium source and NH_3 were separately transported before been merged at around the substrate. The horizontal quartz tube reaction chamber was initially evacuated to a base pressure of 2×10^{-3} Torr (1 Torr \approx 133 Pa) to repel the residual oxygen and introduced into a constant atmosphere nitrogen flow (8 slm N_2 , slm stands for standard liters per minute) to maintain the CVD reactor at 400 Torr. The furnace was then heated up to 960 °C from room temperature with a heating rate of 40 °C/min. The up-pipe NH_3 flow (1.5 slm) and down-pipe N_2 flow (2.3 slm) were then added. The furnace temperature was maintained for 60 min and then followed by cooling down naturally under flowing gas.

Characterization of the aligned GaN nanobelt arrays:

The surface morphology of the GaN nanobelt arrays was observed by a field emission SEM (JSM 6700F) at an acceleration voltage of 5 or 10 kV. The orientation and crystal structure of as-synthesized GaN nanobelt arrays were characterized by SIEMENS D5000 and Bruker D8 X-ray diffractometers, using Cu $K\alpha$ ($\lambda=0.15406$ nm) radiation at 40 kV and 30 mA in a 2θ range from 20 to 80°. More microstructures and orientation characterizations of the GaN nanostructures were carried out using field emission TEM (Tecnai F20 G2) operating at 200 kV. The TEM samples were prepared using the focus ion beam lift-out method. PL was excited by using a continuous wave (CW) He-Cd laser (325nm). The power is 50 mW and the spot size is 300 μm . The emission spectra were analyzed by a Jobin-Yvon Triax 550 monochromator with a 0.025 nm resolution and detected by a Hamamatsu photomultiplier tube and standard photon counting electronics. Raman measurements were carried out using a HORIBA HR800 micro-Raman system coupled with a high stability microscope. The objective of 100 \times was used to collect the signal in back scattering geometry from a focal spot area on the sample of about 1.5 μm in diameter.

The excitation wavelength of 632.8 nm was provided by the internal He-Ne laser.

Part 2. Morphology of aligned GaN nanobelts

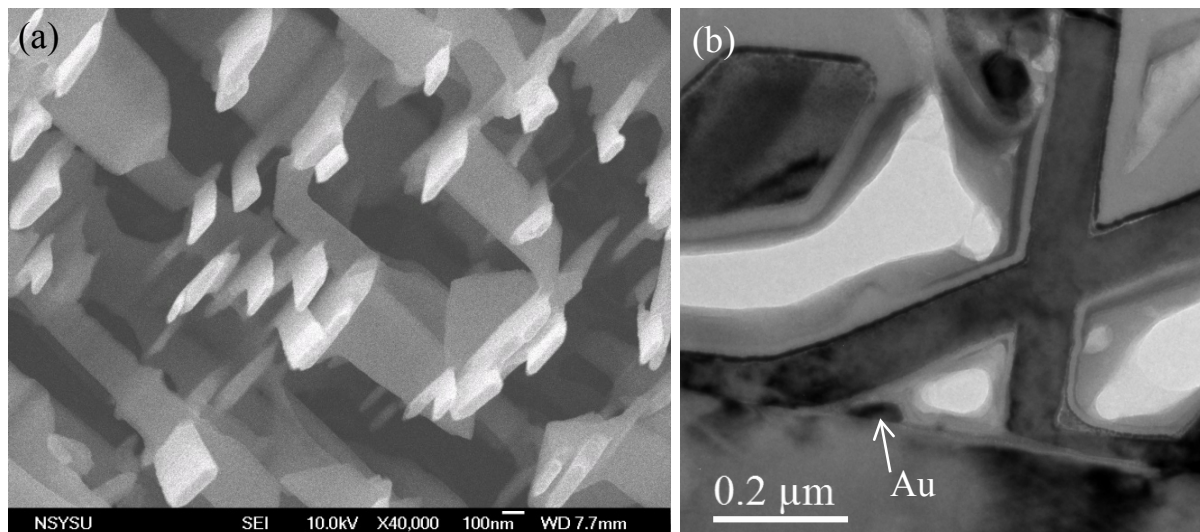


Fig. S1. (a) Top view SEM image and (b) cross-section TEM image of aligned GaN nanobelts grown on (100) γ -LiAlO₂ substrate.

Fig. S1a shows a typical top view SEM image of GaN nanostructure grown on Au-coated (100) γ -LiAlO₂ substrate. Vertical and acclivitous GaN nanobelt arrays are homogeneous distributed on the whole substrate. Fig. S1b shows a cross-section TEM image of the GaN nanobelts taken near the [0002] zone axis. The dark area indicated by the white arrow was determined to be Au by energy dispersive spectroscopy (EDS) analysis.

Part 3. Optical properties of aligned GaN nanobelts

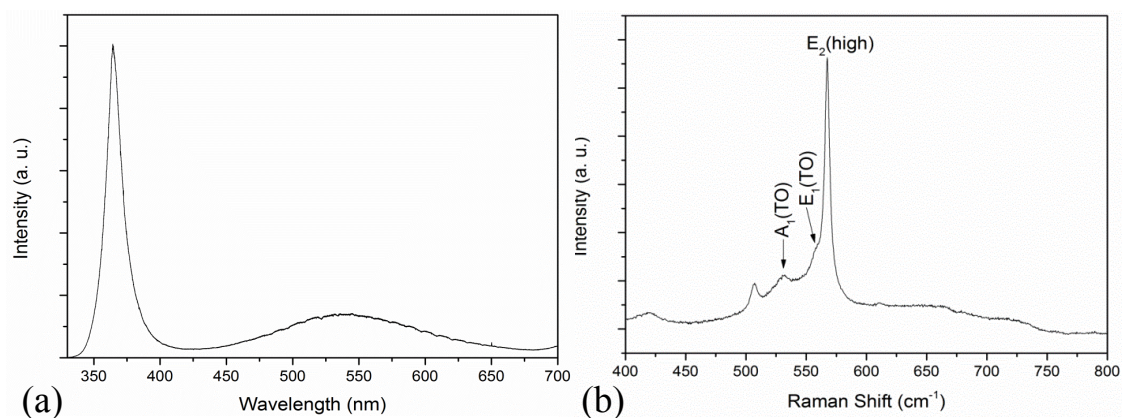


Fig. S2. (a) PL spectrum and (b) Raman spectrum of the GaN nanobelt arrays grown on (100) γ -LiAlO₂ substrate.

Optical properties of as-grown GaN nanobelt arrays were measured by photoluminescence (PL) excited with a 325 nm He-Cd laser beam at room temperature. The typical spectrum recorded from the representative aligned GaN nanobelt arrays (Fig. S2a) exhibits a strong ultraviolet emission centered at around 364.4 nm (3.40 eV) with a full width at half-maximum of 14.0 nm, accompanied a weak, broad yellow band emission at about 539 nm. The ultraviolet PL peak corresponds to the near band edge free exciton emission. The deep-level yellow band emission is due to donor-acceptor recombination in GaN. The intense free exciton peak at room temperature strongly suggests that the nonpolar GaN nanobelt arrays have a high optical quality.

Fig. S2b shows micro-Raman spectrum of the as-grown GaN nanobelt arrays on (100) γ -LiAlO₂ substrate. Apart from the γ -LiAlO₂ substrate phonons, the GaN phonons at $A_1(\text{TO})$: 531.2 cm⁻¹, $E_1(\text{TO})$: 559.0 cm⁻¹, and $E_2(\text{high})$: 567.5 cm⁻¹ are observed. For the wurtzite structure GaN, the $E_2(\text{high})$ phonon frequency is a susceptibility factor of stress. Compressive stress leads to an increase in $E_2(\text{high})$, while tensile stress corresponds to a decrease in $E_2(\text{high})$. The symmetric and strong $E_2(\text{high})$ at 567.5 cm⁻¹ reflects the nearly strainless, high crystalline GaN nanobelt array.

Part 4. Enlarged HRTEM image of Fig. 3d.

