

MBE Self-Assembled Growth of InGaN Nanorods on Patterned Sapphire Substrate with Enhanced Photoluminescence Performance

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Abstract

A controllable self-assembled growth using molecular beam epitaxy (MBE) for dense, uniform, and high-aspect-ratio InGaN nanorods (NRs) with enhanced photoluminescence performance is achieved through lowering Ga/In flux ratio and employing high Miller index planes of patterned sapphire substrate (PSS). This work contributes to deeply understanding MBE self-assembled growth mechanism, as well as controllably fabricating dense, well-separated, and uniform InGaN NRs, thus contributing to the enhanced performance of NR-based devices.

1. Introduction

InGaN NRs exhibiting efficient strain relaxation, large surface/bulk volume ratio, and nearly-free of dislocation, have been employed for NR-based LEDs, solar cells, and photodetectors with enhanced performance owing to its tunable bandgap (0.7–3.4 eV) and high carrier mobility.¹ For achieving the high performance of NR-based device, the uniform, dense, and high-quality NRs are prerequisite. MBE self-assemble growth for InGaN NRs is achieved without using substrate masked with SiO₂/SiN_x nanohole arrays in selective area growth method² and metal catalysts in “vapor-liquid-solid” growth method³. However, a serious problem of MBE self-assembled InGaN NRs is annoying coalescence phenomenon. The NR coalescence induces inhomogeneous strain and dense TDs, leading to undesirable leakage defects and non-radiative recombination centers. Much room is left for systematic studies regarding MBE self-assemble growth of InGaN NRs under varying Ga/In flux ratio. Moreover, PSS, *i.e.*, sapphire substrate with etched pattern arrays comprising of various high index planes with specific surface and structure, offers a huge potential for regulating the uniformity and distribution of spontaneously formed NRs. In this work, we investigated the self-assembled growth of InGaN NRs under varying Ga/In flux ratio on high Miller index planes of PSSs.

2. General Instructions

The Ga/In flux ratio dependence for InGaN NRs on rough C₂-plane of high Miller index ($\{1\bar{1}06\}$) on triangular pyramid PSS was carefully studied. As shown in Fig.1, at the Ga/In flux ratio of 0.37, rough compact layer appeared on C₂-plane. For the Ga/In flux ratio of 0.25, separated nucleated islands and rougher compact layer were formed on C₂-plane. The thickness of compact layer decreased from 157 nm to 124 nm, and the surface roughness significantly increased (Rq , $Rmax=25.79$, 63.48 nm *versus* Rq , $Rmax=13.40$, 36.27 nm).

When the Ga/In flux ratio decreased to be 0.14, separated and inclined NRs were found on rough C₂-plane. At Ga/In flux ratio of 0.05, dense and inclined InGaN NRs appeared on rough C₂-plane. It can be concluded that low Ga/In flux ratio favors 3D growth of NRs instead of 2D film growth.

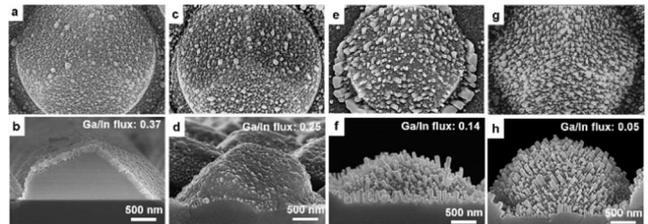


Fig. 1 Top- and side-view SEM images of InGaN compact layers and NRs on rough C₂-plane of triangular pyramid with Ga/In flux ratio of (a) and (b) 0.37, (c) and (d) 0.25, (e) and (f) 0.14, (g) and (h) 0.05.

The Ga/In flux ratio dependence for InGaN NRs on the smooth C₃-plane ($\{4\bar{5}138\}$ plane) of hexagonal pyramid PSS was then carefully explored. As shown in Fig. 2, at the Ga/In flux ratio of 0.37, a rather rough InGaN compact layer constituted by merged NRs was found on C₃-plane. The surface roughness of compact layer on the smooth C₃-plane (Rq , $Rmax=8.76$, 13.32 nm) is smaller than that formed on rough C₂-plane (Rq , $Rmax=13.40$, 36.27 nm). When the Ga/In flux ratio decreased to be 0.25, separated NRs appeared on rougher compact layer. As for the Ga/In flux ratio of 0.14, the dense, obliquely aligned, and highly uniform InGaN NRs can be found on C₃-plane. When the Ga/In flux ratio is reduced to be 0.05, denser, longer, and thinner NRs appeared on C₃-plane.

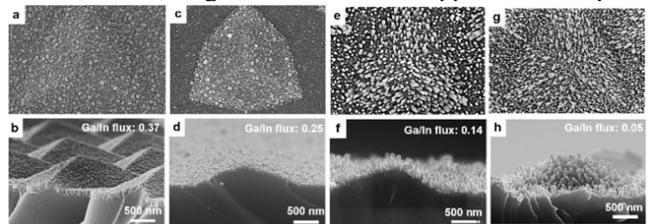


Fig. 2 Top- and side-view SEM images of InGaN compact layers and NRs on smooth C₃-plane of hexagonal pyramid with Ga/In flux ratio of (a) and (b) 0.37, (c) and (d) 0.25, (e) and (f) 0.14, (g) and (h) 0.05.

Using energy dispersive X-ray spectrometer and HRTEM, the component of InGaN NRs with the Ga/In flux ratio of 0.14 and 0.05 are determined to be In_{0.77}Ga_{0.23}N and In_{0.85}Ga_{0.15}N, respectively. For NRs on *c*-plane and C₂-plane, the crystallographic orientation of InGaN NRs is determined to be [0001] through HRTEM and selected area electron diffraction (SAED). Then, we theoretically explore the adsorption energy of Ga and In atoms on side (10 $\bar{1}0$) plane of *c*-orientation InGaN NRs with different Ga content through first-principles

calculations based on density functional theory. E_{ad} values of Ga adatom on (10 $\bar{1}$ 0) plane of In_{0.85}Ga_{0.15}N and In_{0.75}Ga_{0.25}N are calculated to be 7.26 eV and 7.36 eV, respectively. Larger positive value of E_{ad} favors stronger bonding and weaker migration ability of adatom on crystallographic plane,⁴ indicating that higher Ga content of InGaN NRs grown with larger Ga/In flux ratio favors stronger adsorption and lower migration of Ga adatom on (10 $\bar{1}$ 0) plane, leading to faster radial growth of NRs. Moreover, E_{ad} values of In adatom on (10 $\bar{1}$ 0) plane of In_{0.85}Ga_{0.15}N and In_{0.75}Ga_{0.25}N are determined to be 5.44 eV and 5.70 eV, respectively. E_{ad} values of In adatom on (0001) plane of In_{0.85}Ga_{0.15}N and In_{0.75}Ga_{0.25}N are determined to be 9.43 eV and 9.27 eV, respectively. Lower Ga content of InGaN favors weaker adsorption of In adatom on (10 $\bar{1}$ 0) plane, indicating stronger migration ability to top (0001) plane of NRs, leading to intensive axial growth of NRs.

As revealed before, the height and density of NRs grown on various crystallographic planes follow the same order: C₃-plane > C₂-plane > c-plane (Fig. 3a and 3c). By contrast, the diameter of NRs follows the order: C₂-plane > C₃-plane > c-plane (Fig. 3b). As reported, the well-organized step edges in crystallographic planes, as the active sites, facilitate the strong adsorption and ordered nucleation of adatoms⁵. The narrowest step-terrace structures in C₃-plane, *i.e.*, the densest active sites (Fig. 3f), contribute to the highest density of nucleation sites and largest axial growth rate of NRs. For rough C₂-plane (1 $\bar{1}$ 06), NRs on C₂-plane favor larger diameter and smaller density than that on C₃-plane. It can be attributed to the larger size and sparser distribution of the bulges (active nucleation sites) in C₂-plane than that of the narrow step-terrace structure in C₃-plane (Fig. 3e). In addition, c-plane of lowest Miller index with smallest surface free energy, exhibits weakest adsorption, slowest nucleation and migration for InGaN nucleated islands, thus resulting in smallest height, diameter, and density of NRs.

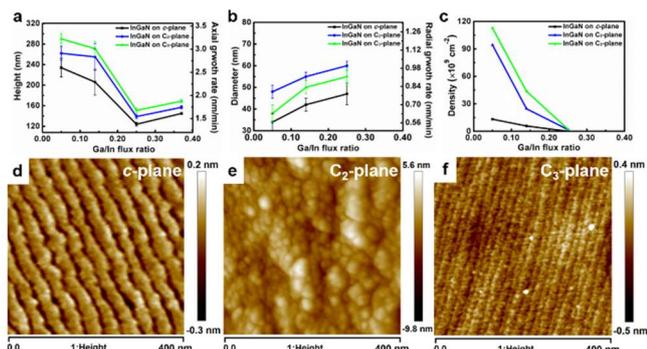


Fig. 3 Evolution of (a) the height and axial growth rate, (b) diameter and radial growth rate, and (c) density of InGaN NRs/compact layers with the Ga/In flux ratio of 0.05, 0.14, 0.25, and 0.37. AFM images of (d) c-plane, (e) the rough C₂-plane, and (f) the smooth C₃-plane.

The crystal orientation of InGaN NRs on c-plane and C₂-plane is accurately determined to be [0001] based on TEM. Then, the interfacial property and epitaxial relationship of InGaN NRs on C₃-plane (4 $\bar{5}$ 1 38) were carefully studied. According to the lattice fringe and inserted SAED pattern (Fig. 4b and 4c), the crystal orientation of NRs on C₃-plane is determined to be [1 $\bar{1}$ 02]. Therefore, the narrow step-terrace

structures in C₃-plane impose strong restriction on self-assembled formation of NRs, leading to the appearance of obliquely aligned NRs with specific orientation.

As shown in Fig. 5, PL intensity of In_{0.77}Ga_{0.22}N NRs on substrates with different patterns and crystallographic planes, *i.e.*, flat c-plane, the rough C₂-plane, and the smooth C₃-plane, follows the order: C₃-plane > C₂-plane > flat c-plane. It is consistent with the order of the total sum of bulk volume and superficial area of NRs grown on these planes over a 1.0 × 1.0 μm² projected area (the PL laser spot size). High index planes on PSS patterns exhibit larger area for nucleation and growth of NRs, and favor larger density and greater aspect ratio of NRs than flat c-plane, leading to superior optical performance.

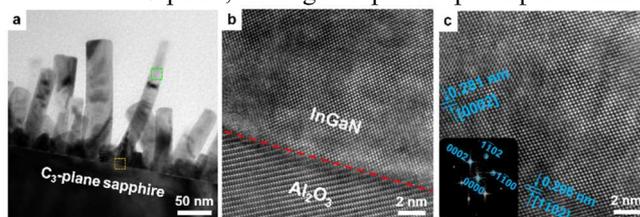


Fig. 4 Cross-sectional TEM image for InGaN NRs on (a) C₃-plane. HRTEM image for (b) InGaN NR/C₃-plane sapphire hetero-interface. HRTEM image for upper zone of NR on (c) C₃-plane and its selected area electron diffraction patterns.

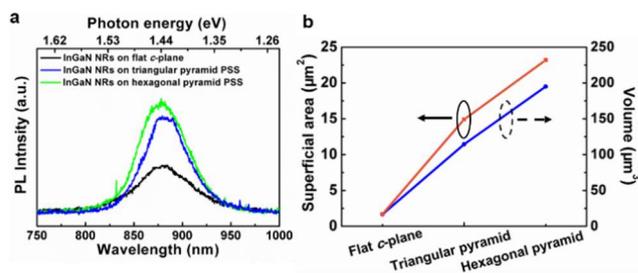


Fig. 5 (a) Room temperature PL spectrum, and (b) superficial area and volume of InGaN NRs (over a 1.0 × 1.0 μm² projected area) with Ga/In flux ratio of 0.14 on flat c-plane, C₂-plane, and C₃-plane.

3. Conclusions

Lower Ga/In flux ratio favors higher density, larger aspect ratio, and smaller coalescence degree of InGaN NRs. High Miller index planes of PSS patterns favor the 3D growth for InGaN NRs with enhanced photoluminescence performance compared with c-plane of low Miller index. The specific surface structure and topography critically affect the morphology, dimension, density, and crystallographic orientation. This work contributes to thoroughly understanding the MBE self-assembled growth mechanism, as well as controllably fabricating dense, well-separated, and uniform NRs, thus facilitating the fabrication of high-performance NR-based device.

References

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