Study of the Indium Content Distribution of Core-shell InGaN/GaN Multi-Quantum Wells (MQWs) on GaN Nanorods

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1. Introduction

Recently, GaN based devices with one-dimensional (1D) nanostructure have gained substantial attention for their potential applications of full color displays and nano-emitters. The one-dimensional structure can be fabricated by top-down patterned etching or bottom-up self assembled growth processes [1]. There are two main issues to grow high performance long-wavelength light emitting diodes (LEDs) and laser diodes with a high indium content. One is the quantum confined stark effect (QCSE) in polar *c*-plane GaN, caused by both spontaneous and piezoelectric polarization Therefore, InGaN/GaN LEDs become very dim in the green emission, this is so-called green gap. Alternatively, growing devices on nonpolar or semipolar planes GaN are adopted to reduced the polarization effect [2]. On the other hand, the large interatomic spacing difference (11%) between InN and GaN results in phase separation. At quite low growth temperature, InGaN films grow with high indium content but poor crystalline quality. The deleterious dislocations lead to strain relaxation when In-GaN films exceed its critical thickness [3]. Semipolar planes, $\{10\overline{1}1\}$, $\{10\overline{1}3\}$ and $\{11\overline{2}2\}$, provide a workable approach to increase the indium content and reduce the polarization related electric fields in the active region of LEDs [4-6].

Our method demonstrates the core-shell InGaN/GaN MQWs grow on GaN nanorods by metalorganic chemical vapor deposition (MOCVD). The core-shell nanorods have the dislocation-free character which can increase internal quantum efficiency (IQE). The 1D nanorods pattern also promotes light extraction efficiency.

2. Experiments

A 2 μ m GaN layer is grown by AXITRON 2000HT MOCVD reactor on a *c*-plane sapphire template. The nanorod arrays are fabricated from a GaN epitaxial wafer by nano imprint patterned etching, followed by epitaxial regrowth. A 0.5 μ m SiO₂ thin film is deposited by plasma-enhanced chemical-vapor deposition and fabricated by nano imprint lithography. The SiO₂ disks are used as masks in inductively coupled plasma reactive ion etching to etch down exposed GaN and form GaN nanorods (Fig. 1(a)), then the disks removed by buffer oxide etch. The GaN nanorod sample is subsequently reloaded in MOCVD rector. The regrowth process forms the crystalline facets on the etched nanorod surfaces. Six pairs of InGaN/GaN MQWs conformally grow on the nanorod crystalline facets at 700 $^{\circ}$ C and 300 torr, as shown in Fig. 1(b)-(c).

The sample is characterized by scanning electron microscopy (SEM), scanning transmission electron microscopy (TEM), energy dispersive spectrometer (EDS) and cathodluminescence (CL) spectroscopy.

3. Results and discussion

The nanorods are fabricated from a GaN substrate by nanoimprint patterned etching, followed by epitaxial regrowth to form crystalline facets. As shown in Fig. 1(c), each arrow-shaped nanorod is composed of a core of GaN nanorods and the shell of InGaN/GaN MQWs. The six side-walls and top-facets are $\{10\overline{1}0\}$ nonpolar planes and $\{10\overline{1}1\}$ semipolar planes, respectively. The cross-section SEM image of nanorods is shown in Fig. 2 (a), an average height of nanorod is about 1.73 µm. The SEM scattering electron detection was switched to CL detection under the same magnification. The spatially integrated CL spectrum of the cross section image is shown in Fig. 2 (b). The first peak around 365 nm is the band edge emission of GaN. The large broad emission peak is from the shell of InGaN/GaN MQWs, and the dominant emission peak is around 500 nm. To reveal the sources of the broad spectrum, the spectrally resolved CL images are shown in Fig. 2 (c)-(i). The MQWs emission wavelength red shift as the location moves from the bottom to top of nanorods.

We further explore the mechanism of wavelength shift from the tip to the bottom of the nanorods. STEM and EDS are manipulated to investigate the structure and composition specifics of the shell of InGaN/GaN MQWs. As shown in Fig. 3 (a), a cross-sectional STEM image viewed in the $[1\overline{1}00]$ zone axis reveal that the InGaN/GaN MQWs conformally grow on GaN nanorods. No dislocation is observed in InGaN/GaN MQWs. A indium content distribution along $\{10\overline{1}1\}$ facets is ~20% at the nanorod tip then falling down to ~15%. A homogeneous thickness distribution of $\{10\overline{1}1\}$ facets indicates that the gradual decreases of indium incorporation efficiency on $\{10\overline{1}1\}$ plane is determined by gas phase diffusion [7]. Of note, asymmetrical thickness distribution for InGaN/GaN MQWs on the two $\{10\overline{1}1\}\$ facets is observed in the STEM image (Fig. 2(a)). Due to $\{10\overline{1}1\}$ planes have an inclination angle of 62° with

c-plane, the thicker right-side inclination plane suggests that a new-born plane is presumably to be the $\{10\overline{1}2\}$ facets (Fig. 3(b)). For the new-born $\{10\overline{1}2\}$ facets on $\{10\overline{1}1\}$ planes supposedly result in the tendency to minimize its total strain energy during growth [8]. According to indium content of MQWs as a function of distance which is perpendicular to the central position of inclination planes on both sides, is described in Fig. 3(c). Indicating that the indium content increases from ~ 8% to ~16% on the left side and ~16% to ~35% on the right side of the inclination surfaces.

The inhomogeneous distribution of indium content on the {1010} plane sidewall is attributed to the decrease of indium diffusion into the bottom portion of nanorod during MQWs growth. However, a high indium content ~33% is observed rounding in the middle of nanorod where is beneath the border region between {1010} and {1011} facets. Owing to the geometric effect of the arrow-shaped nanorods, the narrow spacing of those neighboring nanorods results in indium atoms accumulated in the border region. Compared to the CL spectrum, a indium content ~33% at the middle of nanorod is exactly fit in the position where the dominant emission peak is around 500nm.

4. Conclusions

In summary, core-shell InGaN/GaN MQWs grown on GaN nanorods by MOCVD show the dislocation-free character and a high indium content ~33% in MQWs. A indium content distribution of core-shell MQWs grow on different side facets can be realized by gas phase diffusion, the geometric effect of arrow-shaped nanorods and the strain relaxation mechanism. These results indicate that the 1D core-shell structure have a possibility to replace 2D thin film structure and offer a great potential to fabricate long wavelength GaN-based devices.

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Fig. 1 (a) and (b) is the SEM image viewed in 45° of GaN nanorods and regrowth InGaN/GaN MQWs on GaN nanorods, respectively. (c) The schematic representation of (a) and (b).



Fig. 2 (a) Cross-section SEM image. (b) Spatially integrated CL spectrum of (a). (c)-(i) Spectrally resolved CL images showing the location dependent emission wavelength of quantum wells.



Fig. 3 (a) The cross-sectional STEM and (b) the top view SEM image of a core-shell nanorod. (c) The indium content of MQWs as a function of distance which is perpendicular to the central position of the inclination planes on both sides.

References

- R. Yan, D. Gargas, and P. Yang, Nature photonics 3 (2009) 569.
- [2] S. Nakamura, MRS bulletin 34 (2009) 101.
- [3] S. Keller, B. Keller, D. Kapolnek, U. Mishra, S. DenBaars, I. Shmagin, R. Kolbas, and S. Krishnankutty, J. Cryst. growth 170 (1997) 349.
- [4] R. Sharma, P. Pattison, H. Masui, R. Farrell, T. Baker, B. Haskell, F. Wu, S. DenBaars, J. Speck, and S. Nakamura, Appl. Phys. Lett. 87 (2005) 231110.
- [5] N. Okada, A. Kurisu, K. Murakami, and K. Tadatomo, Appl. Phys. Exp. 2 (2009) 091001.
- [6] H. M. Kim, Y. H. Cho, H. Lee, S. I. Kim, S. R. Ryu, D. Y. Kim, T. W. Kang, and K. S. Chung, Nano Lett. 4 (2004) 1059.
- [7] H. Fang, Z. Yang, Y. Wang, T. Dai, L. Sang, L. Zhao, T. Yu, and G. Zhang, J. Appl. Phys. **103** (2008) 014908.
- [8] Q. Li and G. T. Wang, Appl. Phys. Lett. 97 (2010) 181107.