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Fabricated nano-disk InGaN/GaN multi-quantum well of the inverse hexagonal pyramids

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Abstract

Self-assembled inverted hexagonal pyramids with GaN:Mg and InGaN/GaN multi-quantum-well (MQW) structures were formed through photoelectrochemical wet etching. The formation mechanism of the inverted hexagonal pyramid consisted with a lateral etching process of the InGaN/GaN active layer, bottom-up etching process from N-face GaN direction, and anisotropic etching process. The photoluminescence (PL) intensity of GaN:Mg peak was enhanced in this inverted hexagonal pyramid caused by the quantum confinement effect of this nano-structure. These inverted hexagonal pyramids, consisting of the p-type GaN:Mg, nano-disk InGaN/GaN active layer, and n-type GaN:Si layer, are suitable for nano-scale optoelectronic devices.

1. Introduction

Gallium nitride (GaN), a direct wide band gap semiconductor, has attracted considerable interest in applications for light-emitting diodes (LEDs), detectors, and blue laser diodes. Recently, due to the rapid development of fabrication methods for nano-structures, studies of their optical and electronic properties have attracted a great deal of interest for potential applications in optoelectronic devices, such as quantum cryptography, quantum information, and nano-scale light-emitting devices. To realize the predicted performance of GaN-based nano-scale structures, an intentionally controlled nano-scale structure is needed. Four methods have been used to fabricate GaN-based nano-scale: (1) self-assembled growth quantum dot (QD) (Damilano et al. [1] had reported from visible to white light emission by GaN QDs on Si(1 1 1) substrate); (2) selective growth QD (Tachibana et al. [2] fabricated InGaN QD structures on hexagonal pyramids of GaN, using metalorganic chemical vapor deposition (MOCVD) with selective growth); (3) nano-rod structure, such as inductively coupled plasma-reactive ion etching (ICP-RIE) without a mask [3], synthesis using carbon nano-tubes as templates, [4] growth of single-crystal GaN nano-rods by hydride vapor-phase epitaxy [5]; and (4) self-assembled inverted hexagonal pyramids, reported in this letter.

In a previous report on photoelectrochemical (PEC) etching process on GaN-based material, a roughened surface processed by a laser-lift-off technique followed by an anisotropic etching with a hexagonal "cone-like" surface was reported by Fujii et al. [6] to increase the light extraction efficiency [7].

These inverted hexagonal pyramid structures and the photoluminescence (PL) peak of GaN:Mg layer have stable emission and stronger emission intensity by varying the measurement temperatures. These InGaN/GaN MQW active regions on the pyramids were placed between the top p-type GaN:Mg and bottom n-type GaN:Si layers, and the p–i–n structures have the potential to be applied on the pyramid-shaped light emitters.

2. Experimental

A GaN-based LED structure was grown on 2″ optical-grade C-face (0001) sapphire substrate through a MOCVD

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with a vertical reactor. Precursor gases of trimethylgallium (TMGa), trimethylaluminum (TMAl), trimethylindium (TMIn), and ammonia (NH_3) were used as the Ga, Al, In, and N sources, respectively. The processed LED structure consisted of a 1-μm-thick unintentionally doped GaN layer, a 3-μm-thick n-type GaN:Si layer, six pairs of 3-nm-thick InGaN-well/7-nm-thick GaN-barrier MQW active layers, followed by a 0.1-μm-thick magnesium-doped GaN (GaN:Mg) layer. Prior to the PEC wet-etching process, a mesa region was defined with a depth of about 3 μm by an inductively coupled plasma etcher using Cl_2 gas. During the wet-etching process, 2.2 M potassium hydroxide (KOH) solution was used as the etching solution without any stirring. Along with an illumination exposure from a 400 W Hg lamp for 4 h, an external DC bias fixed at positive 1 V was applied on the n-type GaN:Si layer surface as the anode contact, and platinum was used as the cathode. The etched surface morphologies were observed using JEOL JSM-6700F field-emission scanning electron microscopy (FE-SEM). The PL spectrums were measured by varying the temperature from 12 to 300 K under 325 nm He–Cd laser excitation.

3. Results and discussion

The self-assembled inverted hexagonal pyramids on the mesa region are shown in Fig. 1. The formation mechanism consisted with (1) rapid lateral etching of the MQW layer to form channels attributed to the band gap selective etching; (2) the exposed bottom GaN:Mg N-face surface provided bottom-up etching proceeded vertically along the [0 0 0 1] direction; (3) anisotropic etching occurred with the continuous consumption of the (0 0 0 $\bar{1}$) N-face and the gradual exposure of the {1 0 $\bar{1}$ $\bar{1}$} terminal faces of the GaN:Mg layer; and (4) the formed terminals in a decline constructed an apparently inverted hexagonal pyramid structure. These forming mechanisms for this inverted hexagonal pyramid had been published in our recent work [8]. For some pyramids shown in Fig. 2, the sharp structure was still not obtained, with some remnants of the bottom InGaN/GaN MQW layer due to an incomplete etching process. The six pairs of the InGaN/GaN layers were observed clearly at the bottom of the incomplete inverted hexagonal pyramid structure. The etching rate of InGaN well structure is higher than the GaN barrier structure, and each step profile of MQW structure was seen in one pair of the InGaN/GaN layers. One pair of the InGaN/GaN structure had the decline constructed as the {1 0 $\bar{1}$ $\bar{1}$} terminal faces shown in Fig. 2. After the complete PEC wet-etching process, the inverted hexagonal pyramids structure was determined as a separate inverted hexagonal pyramid unit shown in Fig. 1. Compared with the InGaN QD structures on GaN hexagonal pyramids processed by Tachibana et al. using MOCVD with selective growth, the dimension of the hexagonal pyramids obtained in this experiment was much smaller. The InGaN/GaN MQW layers were formed as a nano-disk active region at the cone tips of the inverted hexagonal pyramids and the radius of the tip of the nano-pyramids was smaller than 10 nm. Moreover, the angle between the top GaN:Mg surface and the pyramid sidewall was measured as around 56.3°, rather close to those reported by Ng et al. [12] and theoretically predicted [9]. From the stacking sequence of Ga and N atoms in GaN with a wurtzite structure, an angle from the top (0 0 0 1) Ga-face plane to the {1 0 $\bar{1}$ $\bar{1}$} terminally inclined faces is clearly identified as 58.4°, confirming our proposed etching mechanism. Because of the crystallographic relationship of 58.4° between the top (0 0 0 1) Ga-face and the {1 0 $\bar{1}$ $\bar{1}$} terminal faces, the size of the pyramids obtained under anisotropic PEC etching was determined by the total thickness of the GaN:Mg and InGaN/GaN MQW layers, i.e. the width of the pyramids was about 1.3 times of the height.

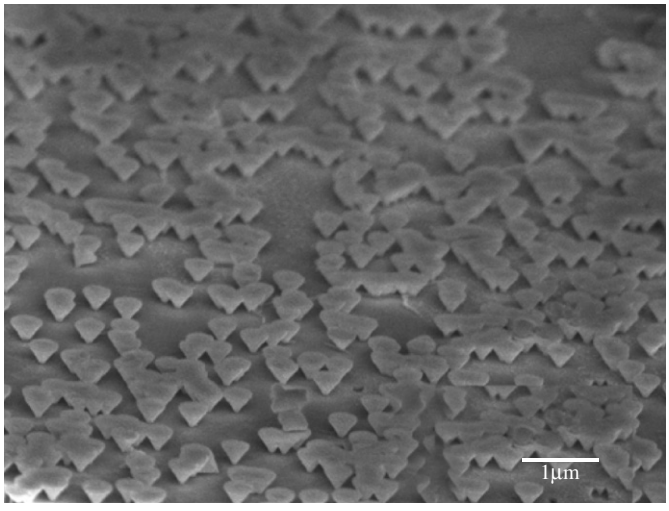

Fig. 1. Self-assembled inverted hexagonal pyramids observed in the mesa region.

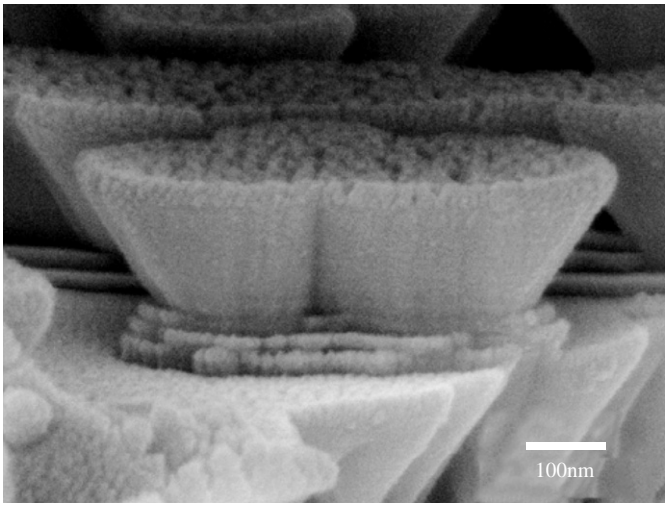

Fig. 2. Self-assembled inverted hexagonal pyramids consisting of the n-type GaN:Si bottom, MQW active layer and top p-type GaN:Mg layer.

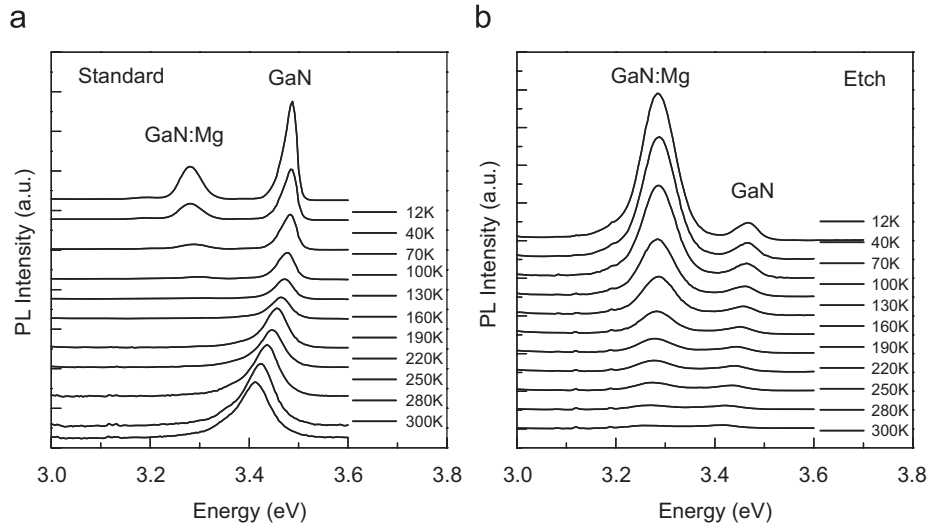

Fig. 3. PL emission spectra of standard (non-etched) and wet-etched LED samples measured at varied temperature.

Fig. 3 shows the PL emission spectra of standard (non-etched) and wet-etched LED samples measured by varying the temperature from 12 to 300 K. For the standard sample, the emission peaks of the GaN band-edge are located at 359.6 nm. By reducing the measurement temperature to 12 K, the GaN:Mg emission peak can be observed at 378.5 nm below 100 K which was caused by thermal quenching [10,11]. After the PEC wet-etching process, the p-type GaN:Mg and InGaN/GaN MQW layers were etched to form the inverted hexagonal pyramid. The emissions peaks of GaN band-edge and GaN:Mg of the PEC-etched sample were found at 360.5 and 378.5 nm. By comparing the intensity ratio of GaN:Mg to GaN band-edge peaks at 12 K, the ratios are 0.33 for standard LED and 8.1 for PEC wet-etching LED structures. After the etching process, the stronger PL emission peak GaN:Mg may be caused by the nano-cavity confined effect and the total reflecting effect of this inverted hexagonal pyramid structure. By varying the measured temperatures, the emission wavelength of GaN band-edge has a blue shift phenomenon. The emission wavelength of GaN:Mg layer had stable emission property around 378.5 nm. This stable emission property of GaN:Mg peak could be caused by the nano-cavity confinement effect in the inverted hexagonal pyramid structure. In Fig. 4, the PL emission peaks of the InGaN/GaN MQW, GaN:Mg, and GaN were both observed at 250 K. The InGaN/GaN MQW peaks were observed at 2.592 and 2.852 eV for standard and etched LED samples. As the nano-disk InGaN/GaN MQW structure was formed, the emission peak was blue shifted from 2.593 to 2.852 eV. The blue shift phenomenon was caused by the strain–relaxed effect and the quantum confinement in this nano-disk MQW structure. The compression strain of the InGaN well structure was caused by the lattice mismatch of the GaN and InGaN epitaxial layer. This nano-disk InGaN/GaN MQW active layer of the inverted hexagonal pyramid structures can partially relax the compressed strain and reduce the piezoelectric effect.

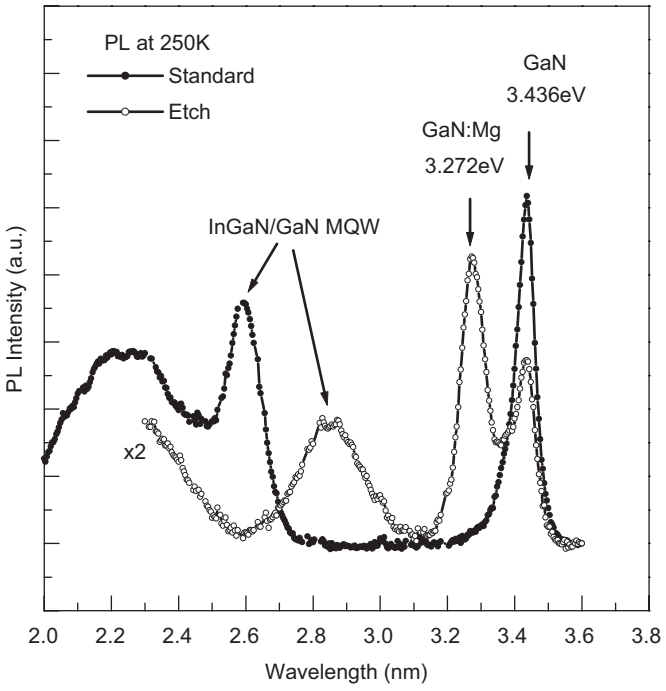

Fig. 4. PL emission spectra of standard (non-etched) and wet-etched LED samples measured at 250 K. The PL emission peaks of InGaN/GaN MQW, GaN:Mg and GaN were both observed.

4. Conclusions

In this paper, a self-assembled inverted hexagonal pyramid consisting with GaN:Mg and InGaN/GaN MQW layers has been successfully formed through a PEC wet-etching process which has 245 nm in width and 184 nm in height. The formation mechanism for the inverted hexagonal pyramid included rapid lateral etching of the InGaN/GaN MQW layer under a band gap selective etching effect, the bottom-up N-face GaN:Mg etching due to high chemical activity, the anisotropic etching of the

GaN:Mg layer with an exposure of $\{1\,0\,\bar{1}\,\bar{1}\}$ terminal faces, and, lastly, the rounding off of the top edges of these pyramids. This stable emission property and higher intensity enhanced ratio of GaN:Mg peak could be caused by the nano-cavity confinement effect in the inverted hexagonal pyramid structure.

Acknowledgments

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References

[1] B. Damilano, N. Grandjean, F. Semond, J. Massies, M. Leroux, Appl. Phys. Lett. 75 (1999) 962.
[2] K. Tachibana, T. Someya, S. Ishida, Y. Arakawa, Appl. Phys. Lett. 76 (2000) 3212.
[3] C.C. Yu, C.F. Chu, J.Y. Tsai, H.W. Huang, T.H. Hsueh, C. F Lin, S.C. Wang, Jpn. J. Appl. Phys. 41 (2002) 910.
[4] W.Q. Han, S.S. Fan, Q.Q. Li, Y.D. Hu, Science 277 (1997) 1287.
[5] H.M. Kim, D.S. Kim, T.W. Kang, Y.H. Cho, K.S. Chung, Appl. Phys. Lett. 81 (2002) 2193.
[6] T. Fujii, Y. Gao, R. Sharma, E.L. Hu, S.P. DenBaars, S. Nakamura, Appl. Phys. Lett. 84 (2004) 855.
[7] E.F. Schubert, Light-Emitting Diodes, Cambridge, UK, 2003.
[8] Chia-Feng Lin, Jing-Jie Dai, Zhong-Jie Yang, Jing-Hui Zheng, Shou-Yi Chang, Self-assembled GaN:Mg inverted hexagonal pyramids formed through a photoelectrochemical wet-etching process, Electrochem. Solid-State Lett. 8 (2005) 12.
[9] J.E. Northrup, L.T. Romano, in: J.H. Edgar, S. Strite, I. Akasaki, H. Amano, C. Wetzel (Eds.), Properties, Processing and Applications of Gallium Nitride and Related Semiconductors, INSPEC, London, UK, 1999, pp. 213–229.
[10] M.A. Reshchikov, G.C. Yi, B.W. Wessels, Phys. Rev. B 59 (1999) 13176.
[11] C. Leey, J.E. Kimy, H.Y. Parky, S.T. Kimz, H. Limx, J. Phys.: Condens. Matter 10 (1998) 11103.
[12] H.M. Ng, N.G. Weimann, A. Chowdhury, J. Appl. Phys. 94 (2003) 650.