



Investigation of GaN-based light emitting diodes with nano-hole patterned sapphire substrate (NHPSS) by nano-imprint lithography

H.W. Huang^{a,b,*}, C.H. Lin^b, J.K. Huang^a, K.Y. Lee^b, C.F. Lin^c, C.C. Yu^b, J.Y. Tsai^b,
R. Hsueh^b, H.C. Kuo^a, S.C. Wang^a

^a Institute of Electro-Optical Engineering, National Chiao-Tung University, Hsinchu 30050, Taiwan, ROC

^b Luxtaltek Corporation, Chunan, Miaoli 350, Taiwan, ROC

^c Department of Material Engineering, National Chung Hsing University, Taichung 400, Taiwan, ROC

ARTICLE INFO

Article history:

Received 22 April 2009

Received in revised form 7 July 2009

Accepted 8 July 2009

Keywords:

GaN

Light emitting diodes (LEDs)

Nano-hole patterned sapphire substrate

(NHPSS)

Nano-imprint lithography (NIL)

ABSTRACT

In this paper, gallium-nitride (GaN)-based light-emitting diodes (LEDs) with nano-hole patterned sapphire (NHPSS) by nano-imprint lithography are fabricated and investigated. At an injection current of 20 mA, the LED with NHPSS increased the light output power of the InGaN/GaN multiple quantum well LEDs by a factor of 1.33, and the wall-plug efficiency is 30% higher at 20 mA indicating that the LED with NHPSS had larger light extraction efficiency. In addition, by examining the radiation patterns, the LED with NHPSS shows stronger light extraction with a wider view angle. These results offer promising potential to enhance the light output powers of commercial light-emitting devices using the technique of nano-imprint lithography.

© 2009 Elsevier B.V. All rights reserved.

1. Introduction

Impressive recent developments of high brightness gallium-nitride (GaN)-based light-emitting diodes (LEDs) have made possible their use in large size flat-panel displays [1,2]. There is still a great need to improve the internal as well as external quantum efficiency to increase their light output power in order to further drive down the total cost of LED modules. Research into improving the light extraction efficiency and brightness in the LEDs [3–8] has been intense. Moreover, high quality GaN-based LEDs have been demonstrated on a micro-scale patterned sapphire substrate (PSS) [7,8], where the micro-scale patterns served as a template for the ELO of GaN and the scattering centers for the guided light. Both the epitaxial crystal quality and the light extraction efficiency were improved by utilizing a micro-scale PSS. Recently, the MOCVD growth of InGaN/GaN LEDs on the PSSs with nano-scale patterns has been reported and compared [9–11]. The LEDs grown on the nano-scale PSS showed more enhancements in the EQE than those grown on the without nano-scale PSS. In this paper, we report a relatively nano-imprinting technique to fabricate a nano-hole patterned sapphire substrate (NHPSS) for mass production. As a result,

the light output efficiency of LED with nano-imprinting (etching depth 165 nm) was significantly higher than that of a conventional LED. Additionally, the intensity–current ($L-I$) measurements demonstrate that the light output power of LED with NHPSS was higher than that of a conventional LED at 20 mA with standard device processing.

2. Experiments

This detailed process flow of nano-imprint lithography on sapphire substrate. Firstly, we spin coated a 200 nm polymer layer on the sapphire sample surface. Secondly, we then placed a patterned mold onto the dried polymer film. By applying a high pressure, we heated the LED samples to above the glass transition temperature of the polymer. Thirdly, The LED samples and the mold were then cooled down to room temperature to release the mold. Finally, we then used a inductively coupled plasma reactive ion etching (ICP-RIE) with BCl_3/Ar plasma to transferred the pattern onto sapphire substrate and remove polymer layer with O_2 plasma etching gas in a reactive ion etching (RIE) system.

Our GaN-based LED samples are grown by metal–organic chemical vapor deposition (MOCVD) with a rotating-disk reactor (Veeco) on a c -axis sapphire (0001) substrate at the growth pressure of 200 mbar. The LEDs structure consist of a 50 nm-thick GaN nucleation layer grown at 500 °C, a 2 μm un-doped GaN buffer, a 2 μm -thick Si-doped GaN buffer layer grown at 1050 °C, an unintentionally doped InGaN/GaN multiple quantum well (MQW) active

* Corresponding author at: Institute of Electro-Optical Engineering, National Chiao-Tung University, Hsinchu 30050, Taiwan, ROC.
Tel.: +886 3 5712121 56327; fax: +886 3 5716631.

E-mail address: steven.huang@luxtaltek.com (H.W. Huang).

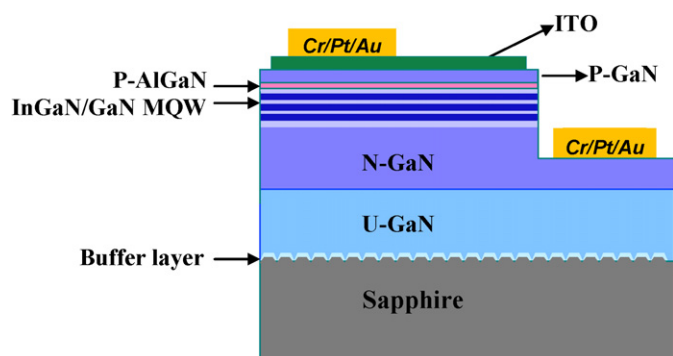


Fig. 1. Schematic diagram of LEDs structure with NHPSS.

region grown at 770 °C, a 50 nm-thick Mg-doped p-AlGaIn electron blocking layer grown at 1050 °C, and a 120 nm-thick Mg-doped p-GaN contact layer grown at 1050 °C. The MQW active region consists of five periods of 3 nm/7 nm-thick $\text{In}_{0.18}\text{Ga}_{0.82}\text{N}/\text{GaN}$ quantum well layers and barrier layers.

The LED samples are fabricated using the following standard processes with a mesa area of $335\ \mu\text{m} \times 335\ \mu\text{m}$. A SiO_2 layer with thickness of 300 nm is deposited onto the LED sample surface by using plasma enhanced chemical vapor deposition (PECVD). Photolithography is used to define the mesa pattern after wet etching of SiO_2 by a BOE solution. The mesa etching is then performed with $\text{Cl}_2/\text{BCl}_3/\text{Ar}$ etching gas in an ICP-RIE system which transferred the mesa pattern onto n-GaN layer. A 270 nm thick ITO layer is subsequently evaporated onto the LED sample surface. The ITO layer has a high electrical conductivity and a high transparency (>95% at 460 nm). Cr/Pt/Au contact is subsequently deposited onto the exposed n- and p-type GaN layers to serve as the n- and p-type electrodes.

Fig. 1 shows the schematic structure of LED with NHPSS. In our study, this type is fabricated in order to investigate the influence of the NHPSS on the LED output power performance. LED structure consists of a Cr/Pt/Au p-electrode, ITO transparent layer, LED epitaxial layers, a smooth p-GaN surface, and a Cr/Pt/Au n-electrode on NHPSS.

Fig. 2(a) top-view and (b) cross-section of SEM images of sapphire with NHPSS. Fig. 2(a) SEM image shows the nano-hole dimension and pattern pitch were approximately 240 and 450 nm. Fig. 2(b) SEM image shows the 12-fold photonic quasi-crystal pattern based on square-triangular lattice (in Fig. 2(b) left-side model). We choose the 12-fold photonic quasi-crystal pattern due to the better enhancement of surface emission was obtained from the PCs with a dodecagonal symmetric quasi-crystal lattice than regular PCs with triangular lattice and 8-fold PQC [12]. The recursive tiling of offspring dodecagons packed with random ensembles of squares and triangles in dilated parent cells forms the lattice. Additionally, in this case we using the dry etching depth and sidewall angle of NHPSS were approximately 165 nm and 45°. We design the 450 nm pattern pitch of PQC in this experiment; the 450 nm pitch is the optimum for the light emission on 460 nm of wavelength.

3. Results and discussion

Fig. 3(a) shows the typical current–voltage (I – V) characteristics. It is found that the measured forward voltages under injection current 20 mA at room temperature for LED with and without NHPSS were 3.11 and 3.08 V, respectively. In addition, the dynamic resistance of LED with and without NHPSS are about 15.9 and 16.0 Ω , respectively. Therefore, in terms of dynamic resistance, there is no influence on this type of devices by incorporating

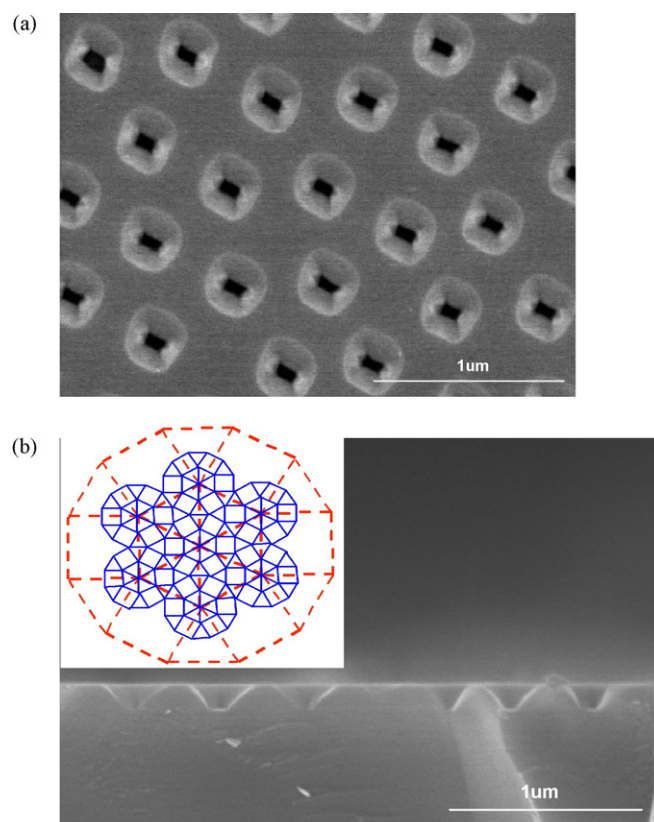


Fig. 2. (a) Top-view and (b) cross-section (inset 12-fold photonic quasi-crystal model) SEM images of sapphire surface with NHPSS.

NHPSS. In Fig. 2(a) the inset EL intensity-current characteristics of conventional LED and LED with NHPSS. The top luminance intensities of the LEDs on wafer were detected using the top-side photo-detectors. It was found that the top-side luminance intensities of the conventional LEDs and LEDs with NHPSS presented data of 70 and 117 mcd at 20 mA, respectively. Therefore, the top-side luminance intensity of the LEDs with NHPSS increased 67% in magnitude as compared with that of the conventional LEDs.

The light output is detected by calibrating an integrating sphere with Si photodiode on the package device, so that light emitted in all directions from the LED can be collected. The intensity–current (L – I) characteristics of the LEDs with and without NHPSS are shown in Fig. 3(b). At an injection current of 20 mA and peak wavelength of 455 nm for TO (Transistor Outline)-can package, the light output powers of LED without NHPSS, and LED with NHPSS (etching depth = 165 nm) on TO-can are given by 14.0 and 18.7 mW, respectively. Hence, the enhancement percentages of LED with NHPSS (etching depth = 165 nm) is 33%, respectively, compared to that of LED without NHPSS. The higher enhancement on standard LED type addresses the effect of the NHPSS which maybe allows the reflect light from sapphire substrate onto the top direction and higher epitaxial crystal quality [9–11] to increase more light output power. If GaN-based LED on sapphire substrate with metallization reflector compared with NHPSS. The simple backside metallization only can increase the EL intensity, but it cannot increase the light output power unless backside reflector of LED with omnidirectional reflector [13]. In addition, the corresponding wall-plug efficiencies (WPE) of conventional LED, LED with NHPSS were 23%, and 30%, respectively, which addresses a substantially improvement by the NHPSS structures as well at a driving current of 20 mA.

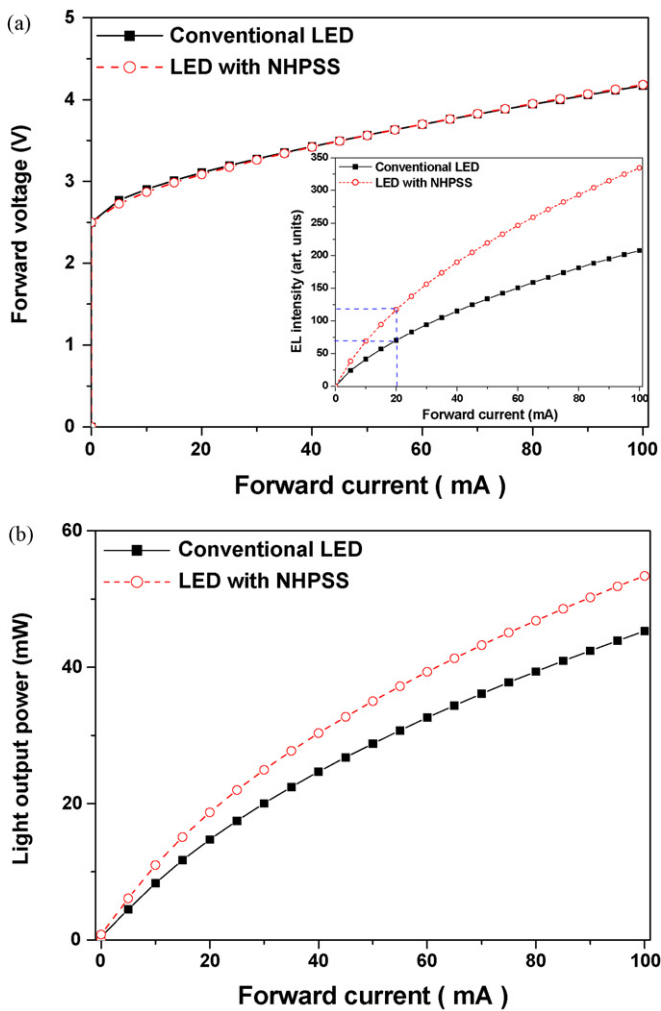


Fig. 3. (a) Measurement current–voltage (I – V) characteristics of LED with/without NHPSS, respectively (inset EL intensity–current characteristics of LED with/without NHPSS). (b) Light output power–current (L – I) characteristics of LED with/without NHPSS, respectively.

Fig. 4(a) shows the integrated PL intensity of InGaN/GaN MQW emission peak with and without NHPSS versus inverse temperature. The temperature dependent photoluminescence (PL) was used to determine the internal quantum efficiency (IQE) of the InGaN/GaN MQW structures. At low temperatures the non-radiative recombination is close to minimum and radiative recombination processes are dominant. With increasing temperature, non-radiative recombination processes get activated and play a key role at room temperature [14]. Fig. 4(a) shows the IQE of LED with and without NHPSS samples calculated from $\text{IQE} = \text{PL}_{300\text{K}} / \text{PL}_{90\text{K}}$; where $\text{PL}_{300\text{K}}$ is the PL intensity at room temperature and $\text{PL}_{90\text{K}}$ is the PL intensity at the lowest temperature in our system. Assuming that the internal quantum efficiency equals unity at 90 K, we obtained the internal quantum efficiency of LED with NHPSS was 34.7% nearly as well as conventional LED 34.4% at RT. Fig. 4(b) shows the SEM image of the cross-section of GaN LED structure with NHPSS. In addition, according to SEM image of cross-section, there is a possible mechanism for forming the voids between GaN and NHPSS. The voids between the sapphire substrate and the GaN layer increase the reflection probability due to scattering at the interfaces of different refractive indices [11]. Thus, a significant amount of photon will be refracted at the GaN/sapphire interface for conventional LED, as shown in Fig. 4(c). Photons generated in the MQW region should experience multiple scattering

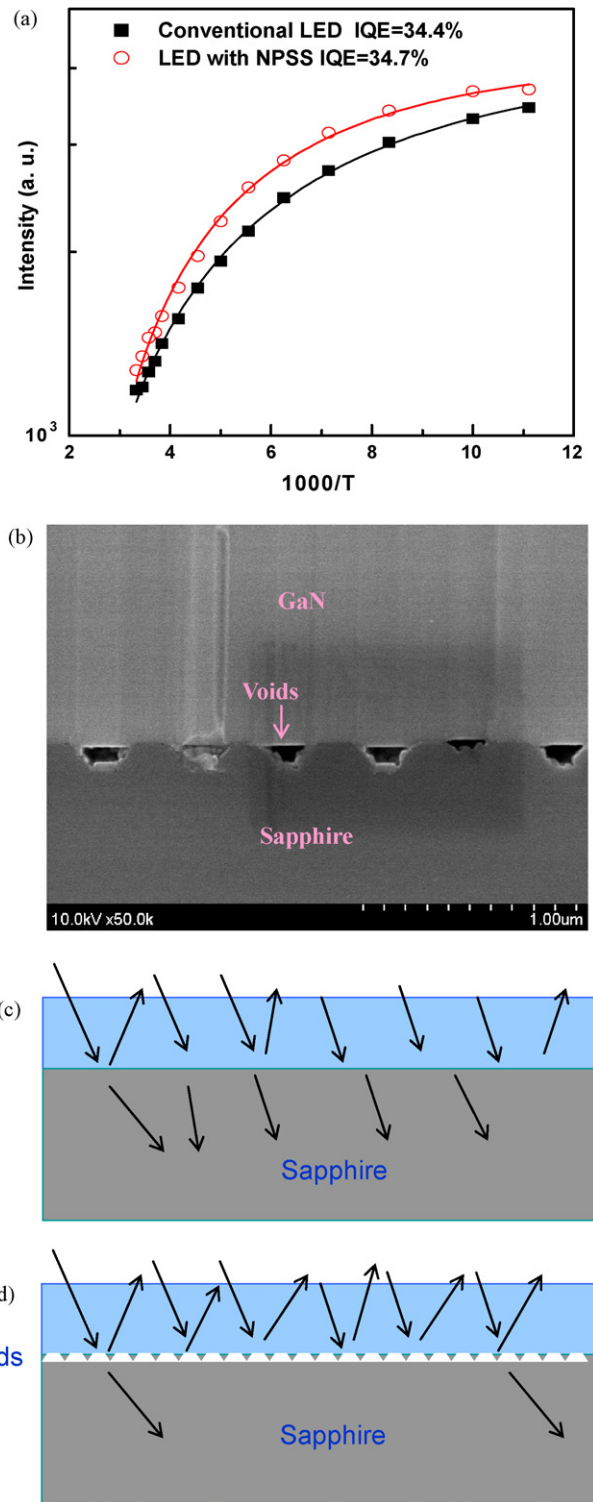


Fig. 4. (a) Arrhenius plots of integrated PL intensities for InGaN/GaN MQW samples, (b) SEM image of GaN LED with NHPSS, (c) schematic drawings of conventional LED and (d) LED with NHPSS.

and reflection at the GaN/sapphire interface of LED with NHPSS sample, as shown in Fig. 4(d). Thus, we can achieve larger output powers from the LEDs with NHPSS. It should be noted that no optimization was performed in the current study. By optimizing the initial pattern on the mold and the etching depth, we should be able to further enhance the LED output power. In this experiment, the LED with NHPSS achieve a light-extraction efficiency

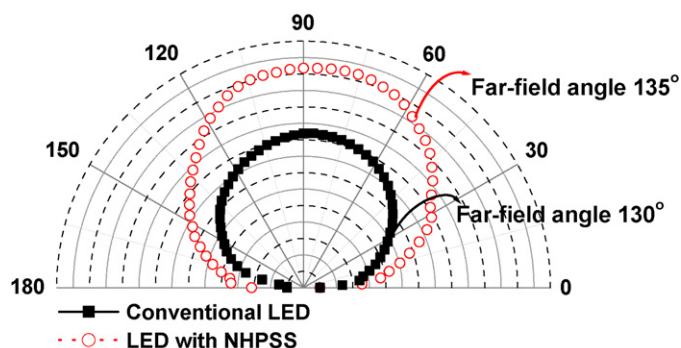


Fig. 5. Far-field patterns of (a) conventional LED and (b) LED with NHPSS structure at driving current 20 mA.

(33%) enhancement of 33% compared to the conventional LED (24%) due to a strongly increase scattering and reflection at the sapphire substrate/GaN interface.

To further study the influence of the PQC structure on the devices, we also measured the light output radiation patterns of the compared LEDs packaged on TO-cans at a driving current of 20 mA, as shown in Fig. 5. It can be seen that the LED with NHPSS structure possesses much higher extraction efficiency with view angle about 135° compared to the view angle of 130° for the conventional LED. This enhancement was attributed to broad light beam shaping effect under NHPSS.

4. Conclusion

In conclusion, GaN-based LEDs with NHPSS by nano-imprint lithography are fabricated and investigated. At a driving current

of 20 mA on TO-can package, the light output power of LEDs with NHPSS (etching depth = 165 nm) is enhanced by a factor of 1.33. The higher output power of the LED with NHPSS is contributed to higher reflectance. The wall-plug efficiency of the InGaN/GaN LED was increased by 30% by NHPSS structure using nano-imprinting lithography. This work offers promising potential to increase output powers of commercial light emitting devices.

References

- [1] M. Koike, N. Shibata, H. Kato, Y. Takahashi, *IEEE J. Select. Top. Quantum Electron.* 8 (2002) 271.
- [2] E.F. Schubert, *Light-Emitting Diodes*, Cambridge University Press, Cambridge, 2003.
- [3] S.J. Chang, C.S. Chang, Y.K. Su, C.T. Lee, W.S. Chen, C.F. Shen, Y.P. Hsu, S.C. Shei, H.M. Lo, *IEEE Trans. Adv. Packag.* 28 (2005) 273.
- [4] J. Song, D.S. Lee, J.S. Kwak, O.H. Nam, Y. Park, T.Y. Seong, *IEEE Photon. Technol. Lett.* 16 (2004) 1450.
- [5] A. David, T. Fujii, B. Moran, S. Nakamura, S.P. DenBaars, C. Weisbush, H. Benisty, *Appl. Phys. Lett.* 88 (2006) 133514.
- [6] J.K. Kim, Th. Gessmann, H. Luo, E.F. Schubert, *Appl. Phys. Lett.* 84 (2004) 4508.
- [7] T.S. Oh, S.H. Kim, T.K. Kim, Y.S. Lee, H. Jeong, G.M. Yang, E.K. Suh, *Jpn. J. Appl. Phys.* 47 (2008) 5333.
- [8] D.S. Wu, W.K. Wang, K.S. Wen, S.C. Huang, S.H. Lin, R.H. Horng, Y.S. Yu, M.H. Pan, *J. Electrochem. Soc.* 153 (2006) G765.
- [9] H. Gao, F. Yan, Y. Zhang, J. Li, Y. Zeng, G. Wang, *J. Appl. Phys.* 103 (2008) 014314.
- [10] J.J. Chen, Y.K. Su, C.L. Lin, S.M. Chen, W.L. Li, C.C. Kao, *IEEE Photon. Technol. Lett.* 20 (2008) 1193.
- [11] C.H. Chiu, H.H. Yen, C.L. Chao, Z.Y. Li, P.C. Yu, H.C. Kuo, T.C. Lu, S.C. Wang, K.M. Lau, S.J. Cheng, *Appl. Phys. Lett.* 93 (2008) 081108.
- [12] Z.S. Zhang, B. Zhang, J. Xu, K. Xu, Z.J. Yang, Z.X. Qin, T.J. Yu, D.P. Yu, *Appl. Phys. Lett.* 88 (2006) 171103.
- [13] C.H. Lin, H.C. Kuo, C.F. Lai, H.W. Huang, K.M. Leung, C.C. Yu, J.R. Lo, *Semicond. Sci. Technol.* 21 (2006) 1513.
- [14] D. Fuhrmann, U. Rossow, C. Netzel, H. Bremers, G. Ade, P. Hinze, A. Hangleiter, *Phys. Stat. Sol. (c)* 3 (2006) 1966.