Displays 32 (2011) 96–99

Contents lists available at ScienceDirect

Displays

journal homepage: www.elsevier.com/locate/displa

Short Communication

Enhanced luminescence of GaN-based light-emitting diodes by selective wet etching of GaN/sapphire interface using direct heteroepitaxy laterally overgrowth technique

Chien-Chih Kao [a], Yan-Kuin Su [a,b,*], Chuing-Liang Lin [c], Jian-Jhong Chen [a]

[a] Institute of Microelectronics, Department of Electrical Engineering and Advanced Optoelectronic Technology Center, National Cheng Kung University, Tainan 701, Taiwan, ROC
[b] Department of Electrical Engineering, Kun-Shan University, Tainan 710, Taiwan, ROC
[c] Department of Electronic Engineering, Kun-Shan University, Tainan 710, Taiwan, ROC

ARTICLE INFO

Article history:
Received 11 May 2010
Received in revised form 20 December 2010
Accepted 27 January 2011
Available online 2 February 2011

Keywords:
Light-emitting diodes (LEDs)
Directly heteroepitaxial lateral overgrowth (DHELO)
Selective wet etching

ABSTRACT

We introduce a method to enhance the luminescence of GaN-based LEDs by combining the direct heteroepitaxy laterally overgrowth (DHELO) technique with selective wet etching process. The epitaxial overgrowth of GaN layers on sapphire substrate with SiO_2 micro-rods array exhibited a reduced dislocation density and improved the crystal quality. The EL intensity of LEDs with SiO_2 micro-rods array was 6.5% higher than conventional LEDs at 20 mA. The selective wet etching process was then used to texture the LED sidewalls into inverted pyramid shape. Finally, the EL intensity could be further enhanced about 12.5% as compared with LEDs with SiO_2 micro-rods array when adopting the textured sidewalls.

1. Introduction

The GaN-based semiconductors have attracted great attention for application in optoelectronic devices such as light-emitting diodes (LEDs) and laser diodes [1]. However, the external quantum efficiency is still low in InGaN/GaN multiple-quantum well (MQW) structures. There are two approaches to increase external quantum efficiency. First, the internal quantum efficiency is strongly influenced by crystal quality and strain-induced piezoelectric polarization [2,3]. Second, the light extraction efficiency of LEDs is limited by total internal reflection due to the large difference in refractive index. However, several approaches have been proposed for reduce the TD density, such as epitaxial lateral overgrowth and patterned sapphire substrate [4,5]. Recently, direct heteroepitaxial lateral overgrown GaN on a SiO_2 or Si_3N_4 patterned sapphire substrate is also a promising way to develop high quality GaN films [6–8]. Furthermore, to improve the light extraction efficiency, surface roughening on p-type GaN surface by dry etching process [9] and sidewalls texturing by wet etching process [10,11] have been widely implemented in GaN-based LEDs. However, it is difficult to etch the Ga-face GaN material due to its chemically stable properties.

In this paper, we designed the SiO_2 micro-rods array to improve the GaN film quality via direct heteroepitaxial lateral overgrowth (DHELO) technique. High quality and low-defect GaN epilayer was lateral overgrown on a SiO_2 micro-rods array patterned sapphire substrate. Moreover, the sidewalls were textured by wet etching process to enhance the light extraction efficiency. The physical, electrical, and optical properties of the LEDs with SiO_2 micro-rods array and textured sidewalls will be discussed.

2. Experimental

Fig. 1a and b shows the schematic diagram of sapphire substrate with SiO_2 micro-rods array at the bottom and surrounding the LED. Firstly, SiO_2 layer was deposited on the sapphire substrate by plasma enhanced chemical vapor deposition. Secondly, the standard photolithography was utilized to define the SiO_2 micro-rods array in the 40-μm-wide isolation which makes the SiO_2 micro-rods array at the bottom and surrounding the LED. The samples were then etched by BOE solution for 2 min of etching time. The diameter, spacing, and height of SiO_2 micro-rods array were 2 μm, 4 μm, and 0.88 μm, respectively. The samples used in this experiment were grown on c-plane (0 0 0 1) sapphire substrate by metal–organic chemical vapor deposition (MOCVD) system. The InGaN/GaN LED structure consists of 30 nm thick GaN buffer

* Corresponding author at: Institute of Microelectronics, Department of Electrical Engineering and Advanced Optoelectronic Technology Center, National Cheng Kung University, Tainan 701, Taiwan, ROC. Tel.: +886 6 2757575 62382; fax: +886 6 235 1864.

E-mail address: yksu@mail.ncku.edu.tw (Y.-K. Su).

0141-9382/$ - see front matter © 2011 Published by Elsevier B.V.
doi:10.1016/j.displa.2011.01.003

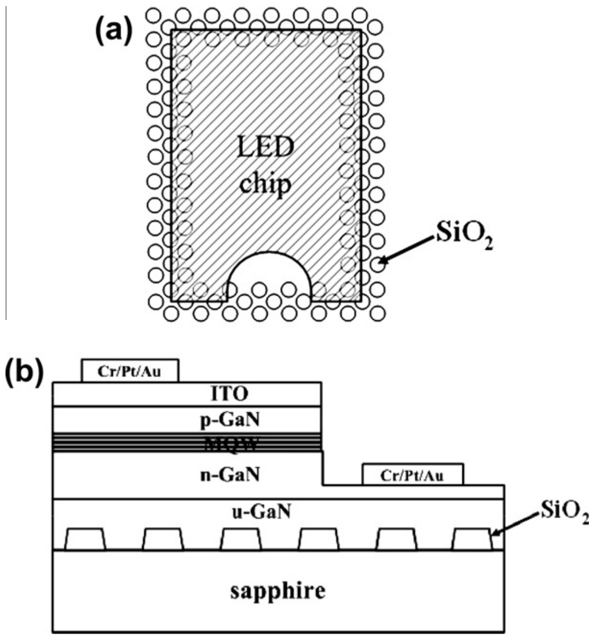

Fig. 1. Schematic diagram of (a) a sapphire substrate with SiO_2 micro-rods array at the bottom and surrounding the LED device and (b) LED structure grown on SiO_2 micro-rods array.

layer grown at 520 °C, a 2 μm thick undoped GaN layer, a 2 μm thick Si-doped n-GaN layer grown at 1050 °C, a 10 nm thick Si-doped n-AlGaN layer grown at 1050 °C, an InGaN/GaN multiple-quantum well (MQW) active region grown at 770 °C, a 40 nm thick Mg-doped p-AlGaN electron blocking layer grown at 950 °C and 0.4 μm thick Mg-doped GaN layer grown at 950 °C. The MQW active region consists of five pairs of 3 nm thick $In_xGa_{1-x}N$ ($x = 0.2$) well layers and 10 nm thick GaN barrier layers. The as-gown wafers were rapidly thermal annealed at 650 °C for 30 min to activate Mg in p-type GaN. The crystalline quality of these samples was analysis by high-resolution X-ray diffraction (HRXRD) and room temperature photoluminescence (PL). The surface morphology and the residual stress were determined by scanning electron microscope (SEM) and micro-Raman spectroscope.

For standard LED processes, we partially etched the samples using Cl_2/Ar as the etching gas by inductively coupled plasma system until the n-type GaN layer was exposed. Indium–tin–oxide was subsequently deposited onto p-type GaN layer to sever as the current spreading layer. Finally, the Cr/Pt/Au deposited on both p- and n-type GaN layer for the metal contact and then separated into 350 μm × 425 μm LED chips from the middle of the isolation. The current–voltage (*I*–*V*) characteristic was measured by Keithley 2400.

3. Results

Fig. 2a and b shows SEM images of p-type GaN grown on sapphire substrate without and with SiO_2 micro-rods array of a device, respectively. The p-type GaN surface morphology was roughened with hexagonal pits for light extraction. It was found that V-shape pit density observed from the area of sapphire substrate with SiO_2 micro-rods array was lower than the area without SiO_2 micro-rods. The lower V-shape pit density was contributed to improved crystalline quality and released compressive strain of the GaN epilayer. As a result, we could change the surface morphology of p-type GaN by changing the design of the SiO_2 micro-rods array.

Fig. 3a shows normalized XRD rocking curves of the (0 0 2) plane measured from these two samples. The crystalline quality could be comprehended by the full width at half maximum

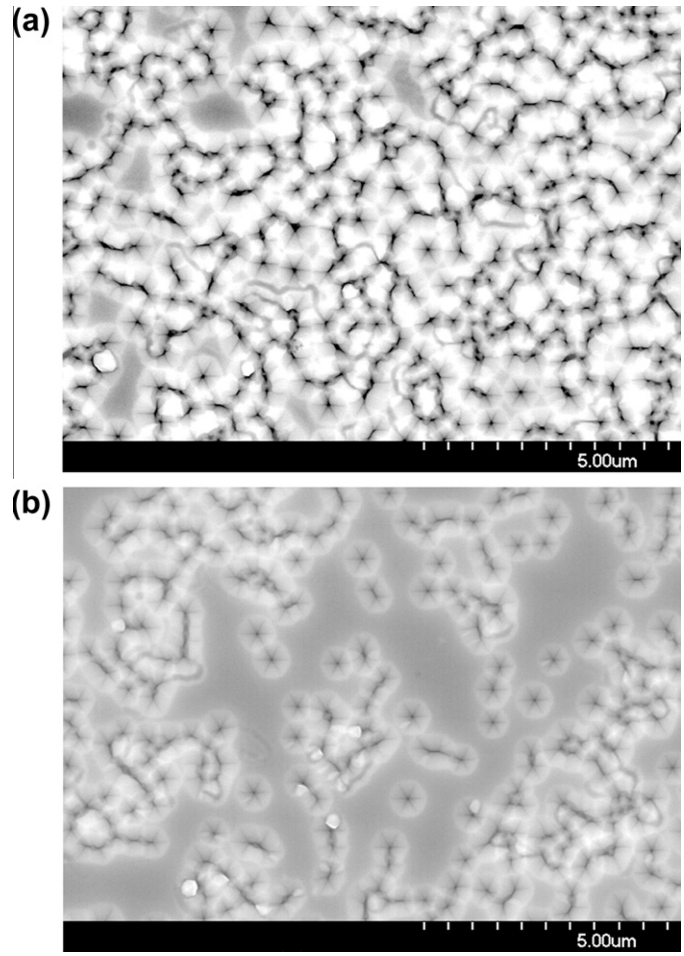

Fig. 2. SEM images of p-type GaN (a) grown on flat sapphire substrate and (b) grown on sapphire substrate with SiO_2 micro-rods array.

(FWHM) of the ω-scan (0 0 2) rocking curve and the etch-pit density (EPD) measurement. The FWHM of the ω-scan rocking curve for the (0 0 2) plane infer the density of screw-type and mix-type dislocations [12]. The FWHM of the conventional LED and LED with SiO_2 micro-rods array were 388.8 arcsec and 313.2 arcsec, respectively. The narrower FWHM implies that crystalline quality could be improved by adopting the SiO_2 micro-rods array. In the EPD measurement, the etching process was carried out in a H_2SO_4 and H_3PO_4 mixture solution with 1:1 ratio at 250 °C for 30 min. The EPD measurement reveals that threading dislocation propagating to the top surface of GaN epilayer which originates from the GaN/sapphire interface. The etch-pit density of the LED with SiO_2 micro-rods array was 1.7×10^{-8} cm^{-2}, which was less than that of conventional LED (4.2×10^{-8} cm^{-2}). The improvement of crystalline quality of GaN epilayer using SiO_2 micro-rods array was attributed to the threading dislocation bending during the lateral overgrowth. Fig. 3b shows normalized room temperature PL spectrum which obtained by using 405 nm GaN laser as excitation light source. The emitting wavelength was blueshift from 445.4 nm to 440.2 nm. The blueshift of the emitting wavelength of the LED with SiO_2 micro-rods array was attributed to smaller residual stress by using the DHELO technique [13]. Fig. 4 shows the E2 (high) mode micro-Raman spectrum of conventional LEDs and LEDs with SiO_2 micro-rods array. The Raman peak position of unstressed GaN bulk [14], LEDs grown without and with SiO_2 micro-rods array were 567.6, 569.426 and 568.508 cm^{-1}, respectively. The Raman peak position shifts to lower wavenumber implies that the residual stress was released during epitaxy lateral overgrowth of GaN epilay-

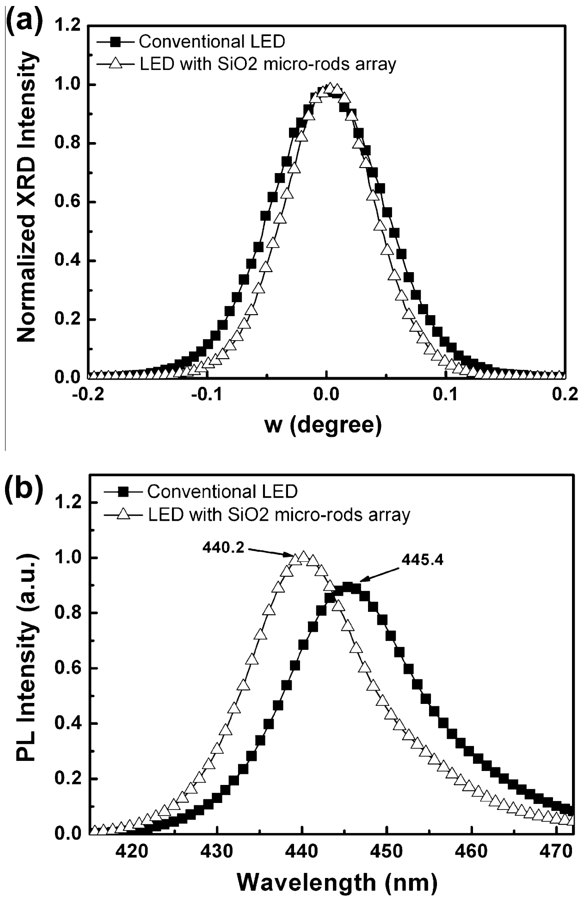

Fig. 3. (a) Normalized XRD rocking curves and (b) Normalized PL spectra measured from the conventional LED and LED with SiO_2 micro-rods array.

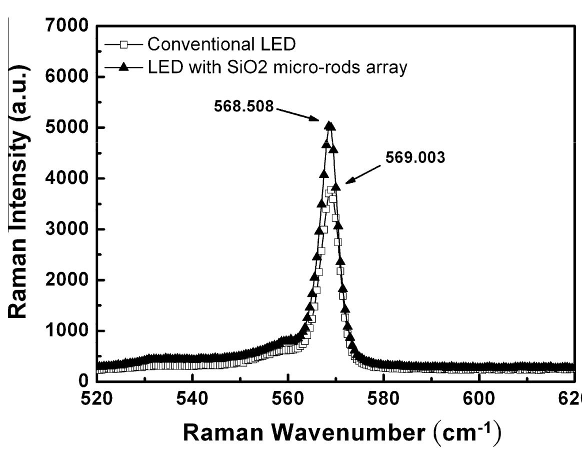

Fig. 4. Micro-Raman spectrum of the conventional LED and LED with SiO_2 micro-rods array.

er. Hence we could estimate the biaxial stress relaxation difference within two samples by the equation: $\Delta\omega = 6.2\sigma_{GaN}$ [15], where $\Delta\omega$ is Raman shift and σ_{GaN} is stress relaxation difference. By analyzing the main scattering signal of E2 (high) mode, the LED with SiO_2 micro-rods array exhibited a 148 MPa (0.918 cm^{-1}) stress relaxation when compared to conventional LEDs. As these results, lattice and thermal expansion mismatch induced residual stress could be released by inserting the SiO_2 micro-rods array. Generally, the strong internal piezoelectric field was found in the GaN epilayer due to high compressive stress. It should be noted that energy band-gap tilts and quantum-confined stark effects (QCSE) caused by the piezoelectric field could also be reduced due to smaller residual stress [16].

For an additional light extraction efficiency improvement, the LED with SiO_2 micro-rods array was further textured GaN/sapphire interface by using selective wet etching process. Firstly, a BOE solution was used to remove SiO_2 micro-rods array to formation of the micro-holes array surrounding the LED. Secondly, a 5M NaOH (50 °C) solution was then used to selective etching GaN/sapphire interface into inverted pyramid shape for 20 min. Fig. 5a and b shows the cross-sectional SEM image of the LED structure grown on SiO_2 micro-rods array and the sample after an etching time of 20 min in 5M NaOH solution at 50 °C. The higher etching rate of the GaN/sapphire interface was observed when the sidewalls of SiO_2 micro-rods array in a NaOH solution. This result was attributed to the two reasons. The first reason is the poor GaN films quality near the edge of the SiO_2 micro-rods array during the lateral overgrowth of GaN films. The second reason of higher etching rate of GaN/sapphire interface is the exposed of N-face GaN after etching in a BOE solution. However, the N-face GaN etches more readily than the Ga-face GaN. The inset of Fig. 6 shows *I–V* characteristics of these three samples. The forward voltages at 20 mA were all approximate 3.3 V for each sample. The similar forward voltage of these samples mean that LED with SiO_2 micro-rods array and additional selective wet etching process will not affect the electrical property. At the reverse bias of 5 V, the leakage current of LED without and with SiO_2 micro-rods array were 24.4 nA and 1.45 nA, respectively. The smaller leakage current observed from the sample can be attributed to better crystalline quality.

Fig. 6 shows the electroluminescence (EL) intensity of the samples as a function of injection current (L-I). The EL intensity of LED with SiO_2 micro-rods array was 6.5% higher than conventional LED at 20 mA. After selective wet etching process of 20 min in a NaOH

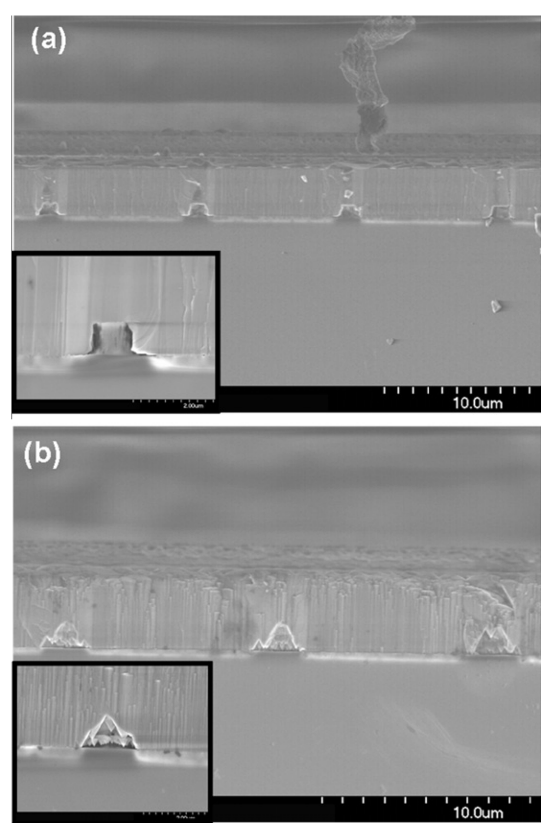

Fig. 5. Cross-sectional SEM image of (a) LED structure grown on SiO_2 micro-rods array and (b) after an etching time of 20 min in 5M NaOH solution.

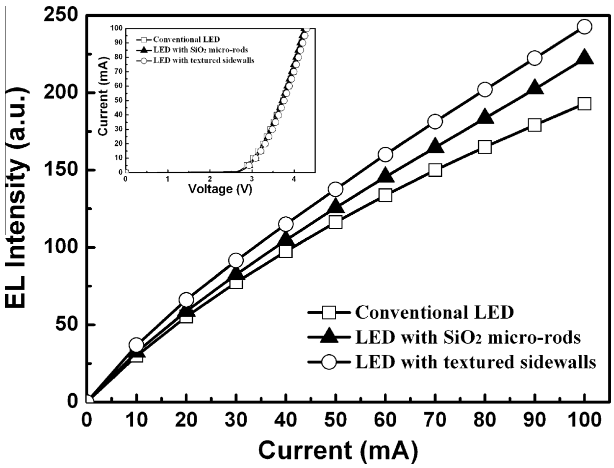

Fig. 6. The EL intensity as a function of injection current for the conventional LED, LED with SiO_2 micro-rods array and LED with textured sidewalls, respectively. The inset shows the forward *I–V* characteristics.

solution, the EL intensity could be further enhanced about 12.5% as compared with LEDs with SiO_2 micro-rods array when adopting the textured sidewalls. In other words, we can achieve significant enhancement in LED luminescence by using simple wet etching process without photo-assistance or high temperature. The improvement was mainly contributed to the trapped light randomly scattered by SiO_2 micro-rods array and textured sidewalls. On the other hand, the epitaxial GaN films quality were also improved by using DHELO technique.

4. Conclusions

In summary, we have demonstrated a method to obtain high performance GaN-based LEDs by combining the DHELO technique with selective wet etching process. However, the lateral overgrowth of GaN layer with the SiO_2 micro-rods array can improve the GaN film quality and release the residual stress. The EL intensity of LEDs with SiO_2 micro-rods array was 6.5% higher than conventional LEDs at 20 mA. Finally, the EL intensity could be further enhanced about 12.5% as compared with LEDs with SiO_2 micro-rods array when adopting the textured sidewalls.

Acknowledgements

The authors would like to thank the Bureau of Energy, Ministry of Economic Affairs, Taiwan, for the financial support under Grant No. 98-D0204-6, and the assistance in devices characterization provided by the LED Lighting and Research Center, NCKU are also appreciated.

References

[1] E.F. Schubert, Light Emitting Diodes, Cambridge Univ. Press, Cambridge, UK, 2003.
[2] Y. Sun, Y.H. Cho, H.M. Kim, T.W. Kang, High efficiency and brightness of blue light emission from dislocation-free InGaN/GaN quantum well nanorod arrays, Appl. Phys. Lett. 87 (2005) 093115.
[3] T. Takeuchi, C. Wetzel, S. Yamaguchi, H. Sakai, H. Amano, I. Akasaki, Y. Kaneko, S. Nakagawa, Y. Yamaoka, N. Yamada, A new bufferlayer for high quality GaN growth by metalorganic vapor phase epitaxy, Appl. Phys. Lett. 72 (1998) 704–706.
[4] A. Sakai, H. Sunakawa, A. Usui, Defect structure in selectively grown GaN films with low threading dislocation density, Appl. Phys. Lett. 71 (1997) 2259–2261.
[5] M. Yamada, T. Mitani, Y. Narukawa, S. Shioji, I. Niki, S. Sonobe, K. Deguchi, M. Sano, T. Mukai, InGaN-based near-ultraviolet and blue-light-emitting diodes with high external quantum efficiency using a patterned sapphire substrate and a mesh electrode, Jpn. J. Appl. Phys. 41 (2002) 1431–1433.
[6] X. Zhang, R.R. Li, P.D. Dapkus, D.H. Rich, Direct lateral epitaxy overgrowth of GaN on sapphire substrates based on a sparse GaN nucleation technique, Appl. Phys. Lett. 77 (2000) 2213–2215.
[7] H.S. Cheong, M.K. Yoo, H.G. Kim, S.J. Bae, C.S. Kim, C.-H. Hong, J.H. Baek, H.J. Kim, Y.M. Yu, H.K. Cho, Direct heteroepitaxial lateral overgrowth of GaN on stripe-patterned sapphire substrates with very thin SiO_2 masks phys, Status Solidi (b). 241 (2004) 2763–2766.
[8] H.G. Kim, T.V. Cuong, M.G. Na, H.K. Kim, H.Y. Kim, J.H. Ryu, C.H. Hong, Improved GaN-based LED light extraction efficiencies via selective MOCVD using peripheral microhole arrays, IEEE Photon. Technol. Lett. 20 (2008) 1284–1286.
[9] D.H. Kim, C.O. Cho, Y.G. Roh, H. Jeon, Y.S. Park, J. Cho, J.S. Im, C. Sone, Y. Park, Won, J. Choi, Q.H. Park, Enhanced light extraction from GaN-based light-emitting diodes with holographically generated two-dimensional photonic crystal patterns, Appl. Phys. Lett. 87 (2005) 203508.
[10] C.F. Lin, Z.J. Yang, B.H. Chin, J.H. Zheng, J.J. Dai, B.C. Shieh, C.C. Chang, Enhanced light output power in InGaN light-emitting diodes by fabricating inclined undercut structure, J. Electrochem. Soc. 153 (2006) G1020–G1024.
[11] D.S. Kuo, S.J. Chang, T.K. Ko, C.F. Shen, S.J. Hon, S.C. Hung, Nitride-based LEDs with phosphoric acid etched undercut sidewalls, IEEE Photon. Technol. Lett. 21 (2009) 510–512.
[12] H. Heinke, V. Kirchner, S. Einfeldt, D. Hommel, X-ray diffraction analysis of the defect structure in epitaxial GaN, Appl. Phys. Lett. 77 (2000) 2145–2147.
[13] C.H. Chiu, Z.Y. Li, C.L. Chao, M.H. Lo, H.C. Kuo, P.C. Yu, T.C. Lu, S.C. Wang, K.M. Lau, S.J. Cheng, Efficiency enhancement of UV/blue light emitting diodes via nanoscaled epitaxial lateral overgrowth of GaN on a SiO_2 nanorod-array patterned sapphire substrate, J. Cryst. Growth 10 (2008) 5170–5174.
[14] V.Yu. Davydov, Yu.E. Kitaev, I.N. Goncharuk, A.N. Smirnov, J. Graul, O. Semchinova, D. Uffmann, M.B. Smirnov, A.P. Mirgorodsky, R.A. Evarestov, Phonon spectrum of wurtzite GaN and AlN experiment and theory, J. Cryst. Growth 189 (1998) 656–660.
[15] T. Kozawa, T. Kachi, H. Kano, H. Nagase, N. Koide, K. Manabe, Thermal stress in GaN epitaxial layers grown on sapphire substrates, J. Appl. Phys. 77 (1995) 4389–4392.
[16] T. Takeuchi, S. Sota, M. Katsuragawa, M. Komori, H. Takeuchi, H. Amano, I. Akasaki, Quantum-confined Stark effect due to piezoelectric fields in GaInN strained quantum wells, Jpn. J. Appl. Phys. 36 (1997) 382–385.