InGaN-based light-emitting diodes with an embedded conical air-voids structure

Yu-Chieh Huang[1], Chia-Feng Lin[1], Sy-Hann Chen[2], Jing-Jie Dai[1], Guei-Miao Wang[1], Kun-Pin Huang[1], Kuei-Ting Chen[1], and Yi-Hsiang Hsu[1]

[1]Department of Materials Science and Engineering, National Chung Hsing University,Taichung, 402, Taiwan
[2]Department of Electrophysics, National Chiayi University, Chiayi, 600, Taiwan
*cflin@dragon.nchu.edu.tw

Abstract: The conical air-void structure of an InGaN light-emitting diode (LEDs) was formed at the GaN/sapphire interface to increase the light extraction efficiency. The fabrication process of the conical air-void structure consisted of a dry process and a crystallographic wet etching process on an undoped GaN layer, followed by a re-growth process for the InGaN LED structure. A higher light output power (1.54 times) and a small divergent angle (120°) were observed, at a 20mA operation current, on the treated LED structure when compared to a standard LED without the conical air-void structure. In this electroluminescence spectrum, the emission intensity and the peak wavelength varied periodically by corresponding to the conical air-void patterns that were measured through a 100nm-optical-aperture fiber probe. The conical air-void structure reduced the compressed strain at the GaN/sapphire interface by inducing the wavelength blueshift phenomenon and the higher internal quantum efficiency of the photoluminescence spectra for the treated LED structure.

OCIS codes: (250.0250) Optoelectronics; (230.3670) Light-emitting diodes.

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1. Introduction

Gallium nitride materials have attracted considerable interest in the development of optoelectronic devices like light-emitting diodes (LEDs), laser diodes, and white LEDs.[1] However, for higher brightness LEDs are required to increase in their internal and external quantum efficiencies. The lower external quantum efficiency of the InGaN-based LEDs is due to a larger refractive index difference between the GaN layer and the surrounding air ($\Delta n \sim 1.5$). Photonic crystal structure formations,[2][3] periodic deflector embedded structures,[4] periodically oriented embedded air protrusion structures,[5] polydimethylsiloxane concave microstructures[6], inverted micropyramid structures,[7] lateral epitaxial overgrowths using pyramidal-shaped SiO_2[8], and roughened N-face GaN surfaces at GaN/Al_2O_3 interfaces[9] have been used to increase light-extraction efficiency in InGaN-based LEDs on Al_2O_3 substrates. Fujii et al.[10] reported that a laser lift-off technique followed by an anisotropic etching process to roughen the surface increases extraction efficiency. It has also been reported that the wet etched undercut sidewalls on the InGaN-based LEDs increase light extraction efficiency.[11][12][13] A chemical lift-off (CLO) technique has been realized by using a CrN layer[14], a ZnO layer[15], and a Si-doped n-GaN layer[16] as sacrificial layers. Most recently, we reported that InGaN-based LED structures grown on triangular-shaped patterned sapphire substrates separated through a chemical lift-off process through laterally etching an AlN sacrificial layer at the GaN/sapphire substrate interface.[17]

In this paper, the conical air-void array of the InGaN-based LED structure was fabricated through dry and crystallographic wet etching processes on an undoped GaN layer, followed by an epitaxial lateral overgrowth (ELOG) process. The ELOG process occurred at the top GaN surface of the truncated conical-hole pattern to form the conical air-void structure. Higher internal quantum and light extraction efficiencies were measured on the treated LED structure. The geometric morphologies, the electrical properties, and the optical properties of the treated LED structures are analyzed in detail.

2. Experiments

The InGaN-based LED wafers were grown on two-sided polished C-face (0001) 2” diameter sapphire substrates through a metal-organic chemical vapor deposition (MOCVD) system. A 30nm low-temperature AlN buffer layer and 4.0μm-thick undoped GaN layer were grown on the sapphire substrate for the following process shown in Fig. 1. Subsequently, a 4μm-thick SiO_2 layer, as a mask layer for the dry and wet etching process, was deposited on the undoped GaN film through a plasma-enhanced chemical vapor deposition (PECVD) system. After a standard photolithographic process and a hydrofluoric acid (HF) wet etching process, the periodic SiO_2 hole patterns, with 3μm-diameter and a 12μm-spacing, were fabricated as shown in Fig. 1(a). In Fig. 1(b), the GaN epitaxial layer was etched to expose the AlN buffer layer through a chlorine-based inductively coupled plasma (ICP) etcher. Then, the wafer was immersed in a hot phosphoric acid (H_3PO_4, 170℃) for an 8min crystallographic wet etching process. The wet etching process consisted of a selective lateral etching process on the AlN buffer layer and a bottom-up N-face crystallographic etching process on the GaN layer where the stable crystallographic etch planes of the GaN layer were formed as shown in Fig. 1(c) and Fig. 2(a). After removing the SiO_2 mask layer, the GaN epitaxial layer was re-grown from the top exposed (0001) GaN facet through a high lateral growth process to coalesce the top surface of the apertures shown in Fig. 1(d) and Fig. 2(c). Then, a 3μm-thick n-type GaN layer, 12 pairs of InGaN/GaN multiple quantum wells (MQWs), and a 0.6μm-thick magnesium-doped p-type GaN layer were grown. The doping source gas for the Si, as the donor, was monosilane (SiH_4) and for Mg as the acceptor, was bis-cyclopentadienylmagnesium (Cp_2Mg). The active layers consisted of a 30Å-thick InGaN-well layer and a 70Å-thick GaN-barrier layer for the InGaN/GaN MQW structure. Next, the n- and p-GaN were defined as the mesa regions through a chlorine-based ICP etcher. The LED chip size was 600×250μm^2 with a 230nm thick indium–tin oxide (ITO) layer deposited on the mesa region as a transparent contact layer (TCL). The Cr/Au metal layers were deposited as n- and p-type contact pads. The LED device that was fabricated with and without the wet etching and overgrowth of the GaN films processes were defined as a conical air-void LED (CAV-LED) and a standard LED (ST-LED). In order to analyze their optical and electrical properties, the chosen ST-LED and CAV-LED devices were located at the 2” LED wafer center. The micro-structures were observed by using a scanning electron microscope (SEM). The optical properties of the LED samples were measured on the basis of photoluminescence (PL) spectra using a 50 mW, 405 nm InGaN laser diode as the excitation source. The PL spectra, electroluminescence (EL) spectra, and light output power were characterized by an optical spectrum analyzer (Ando-6315), an Agilent 4156C precision semiconductor parameter analyzer, and a beam profiler (Spiricon: effective pixels: 1600×1200 pixels), respectively.

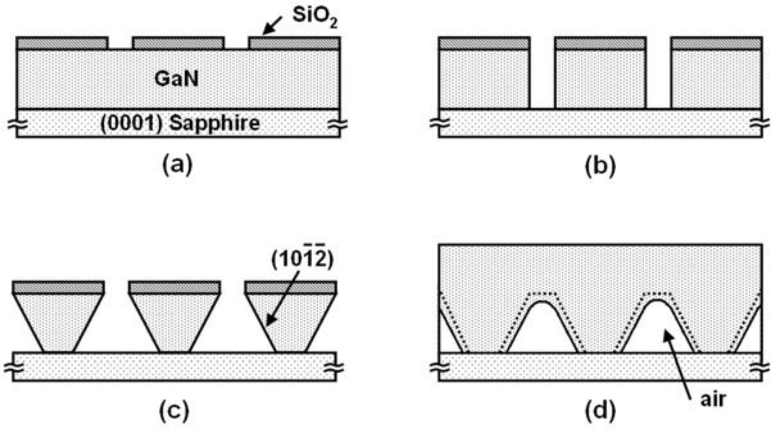

Fig. 1. The fabricated procedures of the conical air-void structure at GaN/sapphire interface are shown.

3. Results and discussion

The SEM micrographs of the GaN undercut structure were fabricated through dry and wet etching processes shown in Fig. 2(a)(b). The stable crystallographic etching planes were observed after the wet etching process that had an including angle with the top GaN (0001) plane calculated as 42°. Stocker et al.[18][19] reported that GaN epilayers have been crystallographically etched as $\{10\overline{12}\}$ planes in a hot H_3PO_4 solution at 132°C. A similar crystallographic etching process to form an inclined sidewall structure has also been discussed in detail in a previous report.[20] The continuous cone-shaped-sidewall was observed at the truncated conical-hole pattern shown in Fig. 2(a). After the crystallographic wet etching process on the undoped GaN layer, the diameters of the top open region and the bottom lateral etched region were measured at 4.7μm and 13μm, respectively. After re-growing the InGaN-based LED structure, the conical air-void structure was observed at the GaN/sapphire substrate interface shown in Fig. 2(c)(d).

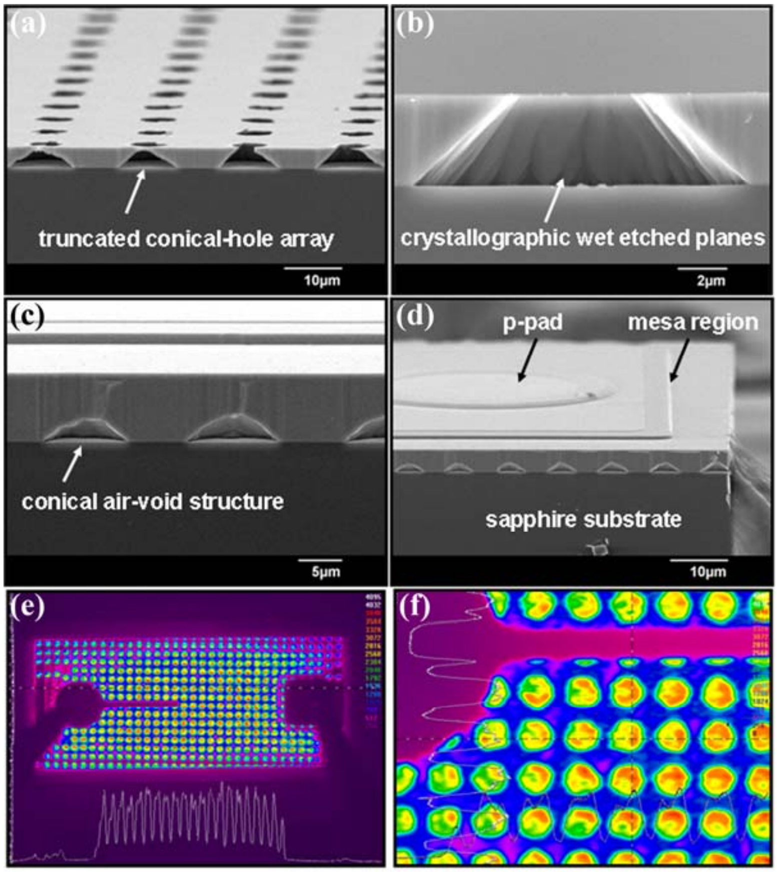

Fig. 2. (a)(b) The SEM micrographs of the truncated conical-hole pattern are observed. (c)(d) After the re-growth process, the conical air-void structures are observed at the GaN/sapphire interface. (e)(f) The light-intensity profiles of the CAV-LED structures were measured at 20 mA. The higher light intensity region is observed as the circular patterns distributed on the whole LED chip.

In Fig. 2(c), the dimensions of the conical air-void structure were measured as a 10μm-diameter-width and a 3.2μm-height. After the ELOG process, the crystallographic etched sidewall of the conical air-void structure became the smooth surface on the cone-shaped structure caused by the lateral overgrowth process that occurred at the top GaN surface of the truncated conical-hole pattern. In Fig. 2(d), the conical air-void structures were observed at the cross-section of the LED chip. The cleaved line was not located on the center of each conical air-void structure so that the width and the height of the conical air-void structure varied depending on the position of the cleaved line. All the CAV structures had smooth cone-shaped sidewall surfaces perhaps caused by the slight lateral growth on the crystallographic sidewall surface. The light-intensity profile of the CAV-LED structure was measured by a beam profiler at a 20mA operating current shown in Figs. 2(e) and 2(f). A higher light intensity region was observed as the circular patterns distributed on the whole LED chip. The order-distributed light-pattern array corresponded to the air-void array on the GaN/sapphire substrate interface. From the top-view light-intensity profile in the Fig. 2(f), the diameter of the high light intensity circular pattern (about 12μm-diameter at the GaN/sapphire interface) was larger than the re-growth conical air void pattern (10μm) caused by a larger light reflection process occurring at the conical sidewall surface of the CAV structure.

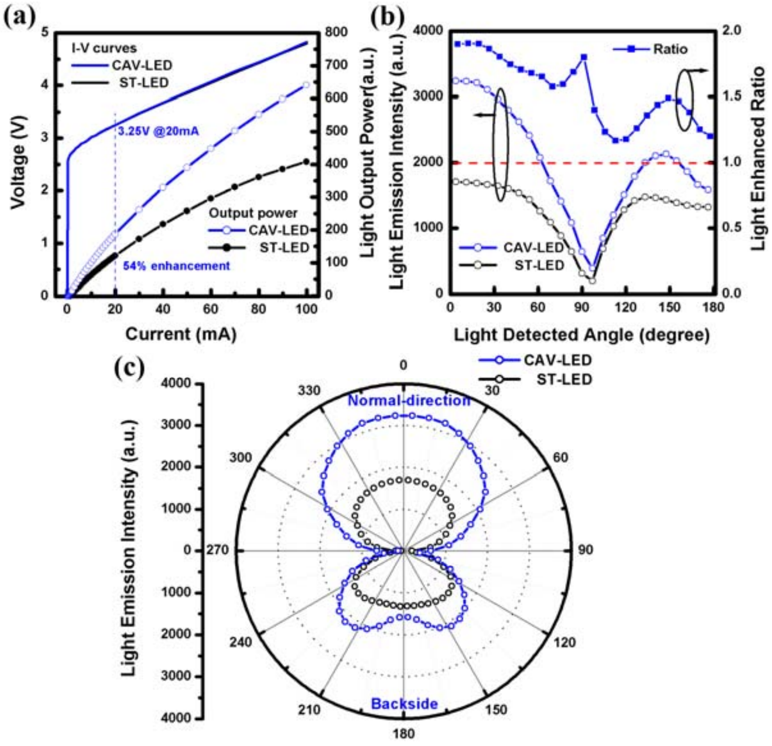

Fig. 3. (a) The light output power and the operation voltage are measured by varying the injection current. (b) The light emission intensity of both LED structures are measured by varying the detected angles from 0^o (normal direction) to 180^o (backside direction). (c) Far-field radiation patterns of both LED structures were measured at 20 mA.

In Fig. 3(a), the operation voltage and the light output power as functions of the injection current were measured. The operation voltages of both LED samples were almost the same at the value of 3.25V at 20mA. The light output power of the CAV-LED had a 54% enhancement compared with the ST-LED at a 20mA operating current. The higher light scattering process occurred at the smooth sidewall of the air-void structure to improve the light extraction efficiency. In the Figs. 3(b)(c), the light-enhanced ratio and the far-field radiation patterns of both LED samples were measured at a 20mA operation current. The detected angles varied from 0° (normal direction) →90° (right side) →180° (back side) → 270° (left side) when analyzing the far-field patterns for both LED samples. The light emission intensity of the CAV-LED structure was higher than that of the ST-LED structure at all detected angles shown in Fig. 3(b). The light-enhanced ratios as a function of the detected angles are defined as the values of the light emission intensity of the CAV-LED divided by the ST-LED at each detected angle by varying from θ=0° (at normal direction) to θ=180°(at backside direction). The highest enhanced ratio of the CAV-LED structure was measured as 1.9 times compared to the one for the ST-LED structure at the normal direction (θ=0°). By summing up the light intensity at all the detected angles, the total light emission intensity of the CAV-LED structure had a 57% enhancement compared to that of the ST-LED structure at a 20mA operating current. In Fig. 3(b), the light enhancement in radiation in the backward direction caused by the light scattering process at the slightly roughened GaN surface at the conical air-void structure and a higher internal quantum efficiency value to generate more emission light from the InGaN active layer. The lower light-enhanced ratio (1.2 times) at 180^{o} detected angle is caused by a partial light reflection occurring at the conical air-void structure that has a GaN/air interface. A divergent angle of an LED is identified as the angle of the half-maximum emission intensity. The divergent angles measured on the front sides of the ST-LED and the CAV-LED were calculated at values of 144^{o} and 120^{o} as shown in Fig. 3(c). The possible reason for the small divergent angles and the higher light intensity for the CVA-LED structure is the emission light from the InGaN active layer scattered to the normal direction by the smooth sidewall around the conical air void structure.

The peak wavelengths of the EL spectra for both LED structures were measured by varying the injection current from 1 to 100 mA as shown in Fig. 4a. The wavelength blueshift phenomenon of the EL spectra was measured as 5.9nm for the ST-LED (from 443.8 to 437.9 nm) and as 5.3nm for the CAV-LED (from 444.1 to 438.8 nm). The wavelength blueshift phenomenon in an InGaN-based LED structure with increasing injection current is due to the band filling effect that increases the injection carriers into the tilted band diagram in the InGaN well structure. The tilted band diagram in the InGaN layer was caused by the compressed strain induced in the piezoelectric field. The smaller wavelength blueshift in the CAV-LED indicated a lower piezoelectric field and a more flattened band diagram in the InGaN well.

The line-scanning EL emission intensity profile was measured by the optical fiber probe that was coated with an Al metal to make an optical aperture. The periodical peak intensity and peak wavelength of the EL spectra were observed at a 0.2mA operation current in Fig. 4(b). The higher and the lower EL intensities had peak wavelengths of 443.8nm at the conical air-void structure and of 444.3nm at the flat GaN/sapphire interface, respectively. In a CAV-LED structure, the short EL emission wavelength was observed at the InGaN active layer above the conical air-void structure that could be caused by the partial release of the compressed strain from the GaN/sapphire interface. The periodical variation of the line-scanning EL peak wavelength was measured as 0.5nm corresponding to the conical air-void structure. During the re-growth process, the lateral overgrowth process occurred at the top GaN surface of the truncated conical-hole structure to improve the epitaxial quality of the top InGaN LED structure. The PL spectra of both LED structures were measured at 300K and 10K as shown in Fig. 4(c) for the ST-LED and 4(d) for the CAV-LED, respectively. At 10K, the peak wavelength and the full width at the half maximum (FWHM) of the PL spectra were measured at 441.5nm (22.6nm) for the ST-LED and at 434.9nm (16.6nm) for the CAV-LED.

The peak wavelength of the laser excitation source was observed at 405nm as the reference wavelength for the PL measurement. The slight peak wavelength blueshift phenomenon and the narrow line width of the PL spectra were observed in the CAV-LED structure. The relative internal quantum efficiencies (integrated PL intensity ratios, I_{300K}/I_{10K}) were calculated at 45% for the ST-LED and at 56% for the CAV-LED structure, respectively. The higher internal quantum efficiency and the narrow PL line width were measured in the CAV-LED structure with the embedded conical air-void structures at the GaN/sapphire interface. From the Fig. 4(b), the slight wavelength blueshift phenomenon of the EL spectrum observed at the conical air-void pattern had a lower piezoelectric field in InGaN active layer. By forming the conical air-voids structures, the CAV-LED structure had a larger light scattering process and a higher internal quantum efficiency when compared to the ST-LED structure.

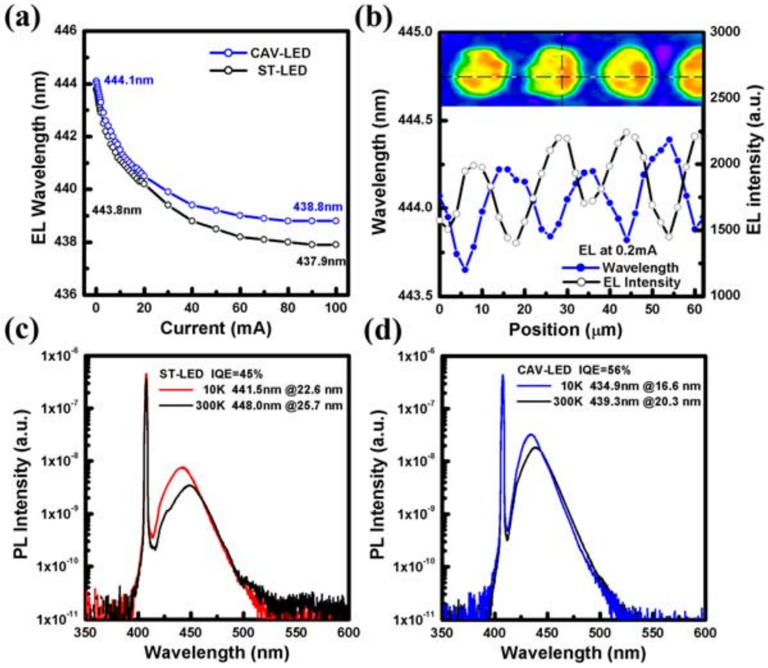

Fig. 4. (a) Peak wavelengths of the EL spectra were measured by varying the operating current. (b) The line-scanning EL emission intensity profile is measured by an optical fiber probe, and the periodical peak intensity and wavelength of the EL spectra are then observed. The PL spectra of the (c) ST-LED and (d) the CAV-LED structures are measured at 10K and 300K to calculate the internal quantum efficiencies.

4. Conclusion

The InGaN LED structure with an embedded conical air-void array at the GaN/sapphire interface has been fabricated. A higher light output power (1.54 times) and a smaller divergent angle (120°) were observed on the CAV-LED structure when compared to the ST-LED structure at 20mA. The additional processes consisted of a dry etching process for 20min process time, a wet etching process for 5min, and an epitaxial regrowth process for the LED structure. In the standard LED, the pattern sapphire substrate was used to increase the light extraction efficiency that the extra cost is added on the pattern sapphire substrate. For the re-growth process, the growth time can be reduced without growing a low-temperature AlN buffer layer and a 4.0μm-thick undoped GaN layer. The higher lateral growth process occurred at the regrowth process to improve the crystal quality and the internal quantum efficiency of the LED structure. A slight PL wavelength blueshift phenomenon was observed at the InGaN active layer above the conical air-void pattern that caused by the reduction of the compressed strain on the GaN/sapphire interface. The higher light extraction efficiency and the higher internal quantum efficiency that were measured at the CAV-LED structure with the conical air-void array have the potential for higher efficiency nitride-based LED applications.

Acknowledgement

The authors gratefully acknowledge the financial support for this research by the National Science Council of Taiwan under grant No. NSC98-2221-E-005-007-MY3 and NSC99-2622-E-005-014-CC3.