Research Article

SiO$_2$ Nanopillars on Microscale Roughened Surface of GaN-Based Light-Emitting Diodes by SILAR-Based Method

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We reported the SiO$_2$ nanopillars on microscale roughened surface on GaN-based LED to enhance light-extraction efficiency. ZnO nanoparticles were deposited on SiO$_2$ as an etching mask before ICP etching SiO$_2$ by successive ionic layer adsorption and reaction method (SILAR), and the different heights of SiO$_2$ nanopillars on microroughened ITO/GaN were obtained after etching. Compared to a regular (flat surface) GaN-based LED, the light output power for a LED with microroughening was increased by 33%. Furthermore, the proposed LEDs with SiO$_2$ nanopillars on microroughened surface show the enhancement in light output power by 42.7%–49.1% at 20 mA. The increase in light output power is mostly attributed to reduction in Fresnel reflection by rough surface. The height of SiO$_2$ nanopillars was increasing cause resulting in more rough on the microscale surface of GaN-based LEDs.

1. Introduction

GaN-based materials and its related ternary compounds such as AlGaN and InGaN have attracted much attention. In the past decades, high-brightness GaN-based LEDs have penetrated the markets of displays, traffic signals, and even solid-state lighting [1–3]. It is required to further extend the application arm of GaN-based LED to projectors automobile headlight and even general lighting; further improvement on optical power and light-extraction efficiency are eagerly required. The reflective index of GaN film is higher than that of air-caused Fresnel reflection. Because of total internal reflection, most of the generated lights in the active layer are absorbed by the electrode at each reflection and gradually disappear. Rough surface is a simple technique and has been used to reduce the Fresnel reflection. Several previous studies that one can increase the light extraction efficiency of GaN-based LEDs by roughening the sample surface have been reported [4, 5], and using ZnO nanopillars to roughen the LED surface retains simplicity and cost effectiveness [6, 7]. Recently, Chang et al. showed that low-temperature (LT) growth conditions could result in many pits on the p-GaN surface to enhance the light-extraction efficiency

[8–11]. The LEDs roughness on the surface showed more enhancements in the EQE than those without roughness on surface. Furthermore, a wide range of chemical and physical deposition techniques have been used to fabricate ZnO films, including reactive and nonreactive sputtering [9], chemical vapor deposition [10], pulse laser ablation [11], sol-gel [12], spray pyrolysis [13], chemical bath deposition [14], and SILAR [15]. Among these, the solution approach based on soft chemical technique has attracted increasing attention in recent years [16], due to its high reliability, low cost, and large-area deposition compared with other methods. In addition, the resulting light output power efficiency of these LEDs is significantly higher than that of conventional LEDs. In this paper, we reported the SiO$_2$ nanopillars on roughened surface on GaN-based LEDs to enhance light extraction efficiency. The SiO$_2$ film was prepared and controls the thickness by using plasma-enhanced vapor-phase epitaxy (PECVD), and ZnO nanoparticles which we can control the density is prepared as a mask by SILAR [17]. Then, SiO$_2$ layer was etched by ICP to get the different height of SiO$_2$ nanopillars on the ITO surface. As a result, the light output power efficiency of LED with SiO$_2$ nanopillars was significantly higher than that of a conventional LED.
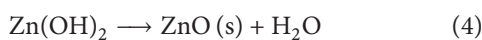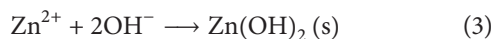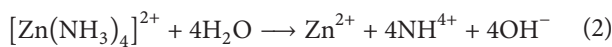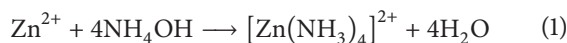
2. Experimental

The GaN-based LED structure was grown on c-axis sapphire substrates by using low-pressure metal-organic chemical-vapor deposition. N-type GaN epitaxial layers, which were including a $1\,\mu$m thick undoped GaN layer and a $2\,\mu$m thick Si-doped n-GaN layer, were fabricated on sapphire substrates as templates for the subsequent regrowth process, before the growth of LED structures. The five periods InGaN/GaN multiple quantum well (MQW) with emission wavelength in the blue region, a 50 nm thick Mg-doped p-AlGaN electron blocking layer, and a $0.2\,\mu$m thick Mg-doped p-GaN cladding layer were grown at 1050°C. After these grown layers, the 500 nm thickness of low-temperature Mg-doped p-GaN cap layer was grown at 770°C. Finally, a heavy 5 nm thick Si-doped n+-GaN tunnel layer was grown on the low-temperature p-GaN cap layer to make the ohmic contact with the ITO transparent contact layer (TCL) [18]. The LEDs had a mesa structure with an area of $300 \times 300\,\mu$m^2. Before fabricating the LEDs with SiO$_2$, a 3200 nm thick ITO film was deposited on the top of GaN-based LEDs by radio frequency magnetron sputtering, and then we roughed the surface by dipping ITO etchants for several seconds. Cr/Au layers as the p- and n-contact electrodes were fabricated. A different thickness 200 nm–600 nm of SiO$_2$ film was deposited on an ITO layer by PECVD. The photoresist layer covered the metal contacts by photolithography in order to prevent the SiO$_2$ growing on the ohmic contact electrodes. The ZnO nanoparticles were grown on the top of SiO$_2$ film as the etching mask in 6 cycles by SILAR. The detailed procedures of ZnO nanoparticles in one cycle are shown in Figure 1 and described as follows.

The rinsing procedure by SILAR method:

(a) dipping glass substrates in the zinc complex [Zn(NH$_3$)$_4$]$^{2+}$ solution for 20 s,

(b) Zn(OH)$_2$ precipitation is formed on the substrates by dipping in unheated DI water for 20 s,

(c) dipping glass substrates in ultrasonic-assisted DI water for 30 s to remove counterion Cl$^-$ and loosely attached Zn(OH)$_2$ grains,

(d) dipping glass substrates in 95°C DI water : ethylene glycol = 1 : 4 for 20 s to form ZnO nanoparticles,

(e) keeping substrates in ultrasonic-assisted DI water for 30 s to remove the loosely attached ZnO and unreacted Zn(OH)$_2$ grains.

The relative reactions were shown as the following equations:

$$Zn^{2+} + 4NH_4OH \longrightarrow [Zn(NH_3)_4]^{2+} + 4H_2O \qquad (1)$$

$$[Zn(NH_3)_4]^{2+} + 4H_2O \longrightarrow Zn^{2+} + 4NH^{4+} + 4OH^- \qquad (2)$$

$$Zn^{2+} + 2OH^- \longrightarrow Zn(OH)_2\,(s) \qquad (3)$$

$$Zn(OH)_2 \longrightarrow ZnO\,(s) + H_2O \qquad (4)$$

After the deposition of ZnO nanoparticles on SiO$_2$ layer, we sequentially etched SiO$_2$ by using ICP with ZnO nanoparticles as the etching nanomasks. We then get the different heights of SiO$_2$ nanopillars on an ITO film. Finally, we removed the ZnO nanoparticles and photoresist by HCL and acetone. A schematic diagram of GaN-based LED with SiO$_2$ nanopillars is shown in Figure 2. The characteristics of current-voltage (I-V) and current-power were measured at room temperature using Keithley 2430 source meter combined with an integrating sphere and a spectrum meter.

3. Results and Discussion

Figure 3 shows the tiled-view and cross-section SEM images of the microrough surface and after SiO$_2$ etching by ICP in different height of SiO$_2$ nanopillars on rough p-GaN surface: Figure 3(a) showed the image of surface of ITO on microrough p-GaN and named LED II; Figures 3(b) and 3(e) showed the images of 200 nm SiO$_2$ nanopillars on ITO, and named LED III; Figures 3(c) and 3(f) showed the images of 400 nm SiO$_2$ nanopillars on ITO and named LED IV; Figures 3(d) and 3(g) showed the images of 600 nm SiO$_2$ nanopillars on ITO and named LED V. The conventional GaN-based LED with flat surface was named LED I. After the ZnO nanoparticles were deposited at 95°C in the solution with ratio of DI water : ethylene glycol = 1 : 4 and SiO$_2$ by etching by ICP, Figure 3(b) shows that SiO$_2$ columns were from 180 to 250 nm and the spacings were approximately 100 nm to 200 nm. Figure 3(c) shows that SiO$_2$ columns were from 150 to 200 nm, and the spacing was approximately 100 nm to 200 nm. Figure 3(d) shows that SiO$_2$ columns were from 100 to 150 nm, and the spacing was approximately 100 nm to 200 nm. The diameter of SiO$_2$ columns reduced as the etching time longer because of side-etching by ICP.

Figure 4 shows the current-voltage (I-V) characteristics of GaN-based LEDs with and without SiO$_2$ nanopillars. At a current of 20 mA injection, it was found that forward voltages were 3.19, 3.22, 3.21, 3.22, and 3.22 for LED I, LED II, LED III, LED IV, and LED V, respectively. It showed that the forward voltages of GaN-based LEDs plane and microrough surface with and without SiO$_2$ nanopillars were very similar I-V characteristics. This result indicates that the electrical properties of the ITO film and MQWs do not degrade the electrical properties after etching SiO$_2$ process, because of using low ICP power.

Figure 5 demonstrates light output power of GaN-based LEDs with and without SiO$_2$ nanopillars at a driving current of 20 mA. The output intensities of these LEDs increased with injection current when the injection current was small can be seen. Furthermore, it was found that output powers observed from the four LEDs with SiO$_2$ nanopillars were all larger than that observed from the conventional LED again. With 20 mA injection current, it was found that output powers of these LEDs were 6.36, 8.45, 9.08, 9.35, and 9.48 mW for the LED I, LED II, LED III, LED IV, and LED V, respectively. It was shown that smaller output power of LED I is attributed to Fresnel reflection. In other words, we can enhance the light output power at 20 mA by 33% by roughened p-GaN attributed to low-temperature growth for LED II. Furthermore, we can enhance the light output power at 20 mA by
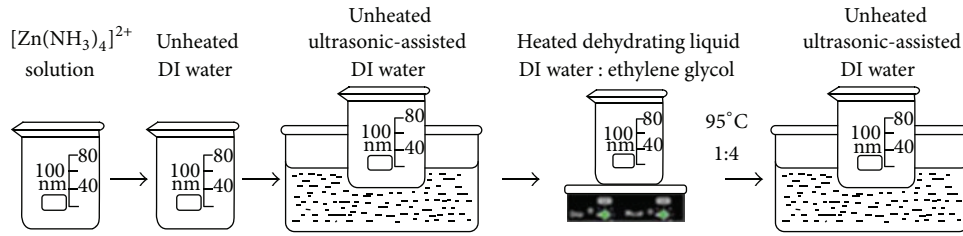
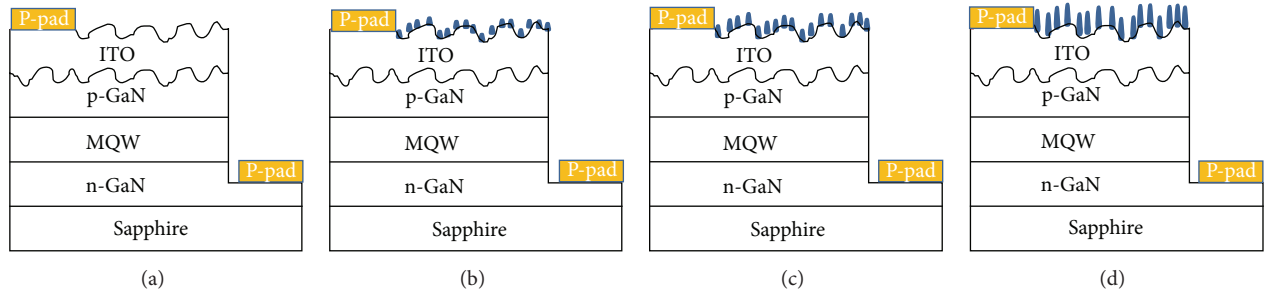FIGURE 1: The detailed procedures of ZnO nanoparticles in one cycle.

FIGURE 2: Schematic diagram of GaN-based LEDs with different heights of SiO_2 nanopillars: (a) without SO_2 nanopillars; (b) 200 nm; (c) 400 nm; and (d) 600 nm.
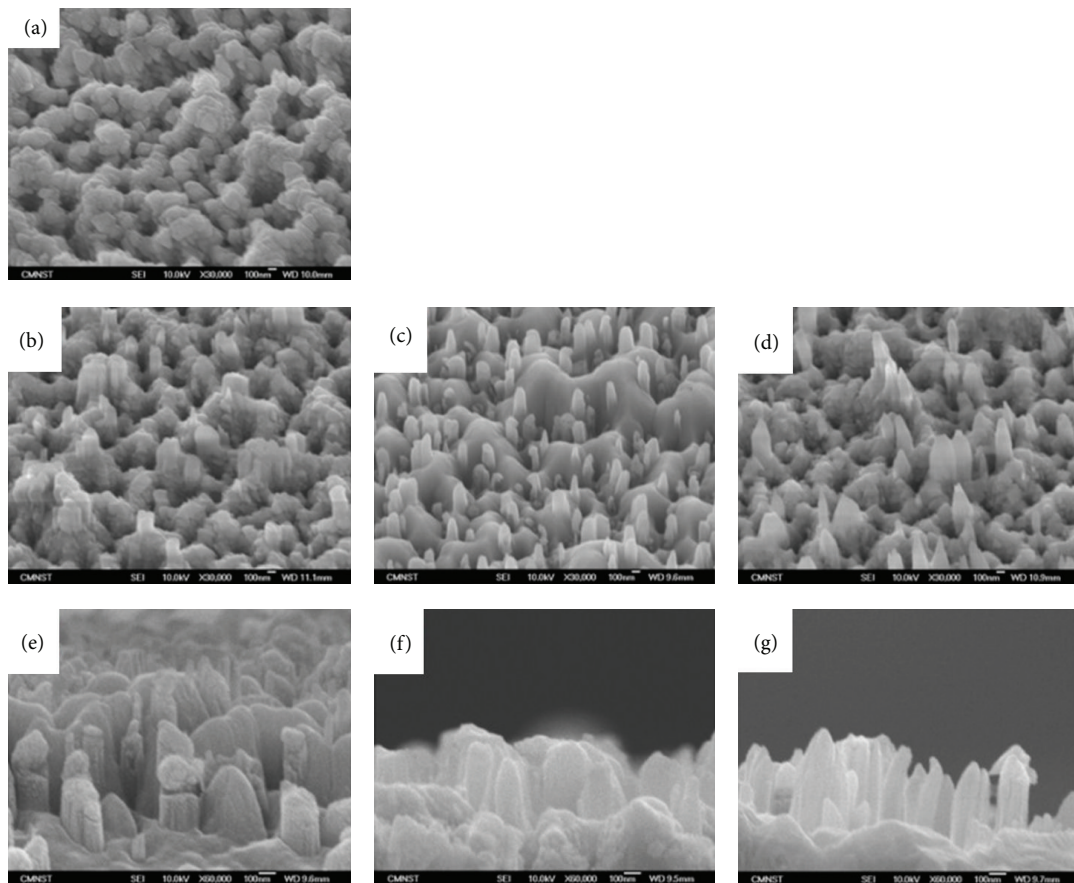

FIGURE 3: Tilted-view and cross-sectional SEM images of the surface after etching SiO_2 by ICP in various heights of SiO_2 nanopillars: (a) roughened surface of ITO on p-GaN; (b) and (e) 200 nm SiO_2 nanopillars: LED II; (c) and (f) 400 nm SiO_2 nanopillars: LED III; and (d) and (g) 600 nm SiO_2 nanopillars: LED IV.
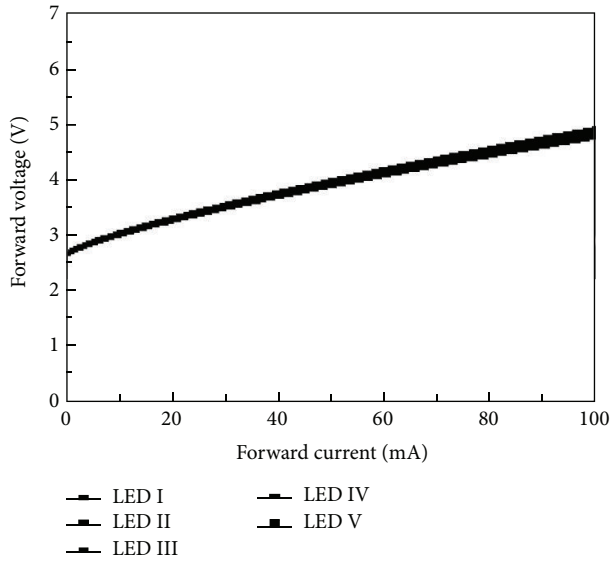
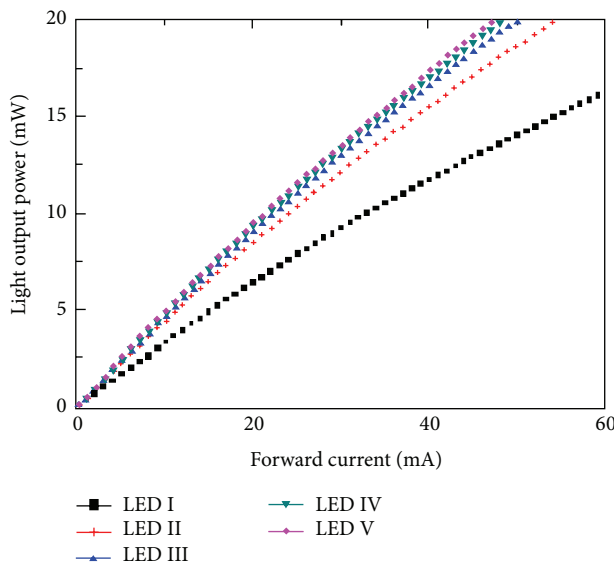FIGURE 4: *I-V* curves of GaN-based LEDs with and without SiO$_2$ nanopillars.

FIGURE 6: EL spectra of GaN-based LEDs with and without SiO$_2$ nanopillars.

FIGURE 5: Light output power versus current characteristics of GaN-based LEDs with and without SiO$_2$ nanopillars.

nanopillars on microrough surface of ITO was decreased. The diameter of SiO$_2$ nanopillars gets smaller after ICP etching.

Figure 6 presents the room-temperature electroluminescence (EL) spectra of the flat-surface LED and LEDs with SiO$_2$ nanopillars at a driving current of 20 mA. The GaN-based MQW emission peaks of the five devices were observed at 475 nm. The EL intensity of conventional LED I was the lowest, while the intensities of LED II to LED V were noticeably higher. These results may be attributed to superior light extraction efficiency resulting from the nanopillars structure, which was further improved by roughening the surface of the LEDs. It was also found that EL intensities observed from the LEDs with SiO$_2$ nanopillars on the surface were all larger than those observed from the conventional LEDs. This could be attributed to the enhanced light extraction efficiency of the textured surface by SiO$_2$ nanopillars. Additionally, it was found that EL intensity increased with increasing the density of SiO$_2$ nanopillars. Such a result suggests that nanosized SiO$_2$ nanopillars did scatter and enhance the light output more effectively.

4. Conclusion

In summary, we have successfully demonstrated a feasible method to enhance light extraction by producing SiO$_2$ nanopillars on LED surfaces by solution method. By using ZnO nanoparticles as a dry etching mask on SiO$_2$ film before SiO$_2$ etching by ICP. By this method, we can control the density and diameter of ZnO nanoparticles. The enhancement of light output power of the GaN-based LEDs with SiO$_2$ nanopillars is achieved up to 42.7%–49.1% compared to that of the conventional LED at injection current of 20 mA. The enhancement is attributed to the reduction of Fresnel reflection. The height of SiO$_2$ nanopillars was increased due

7.5%, 10.6%, and 11.2% with different height SiO$_2$ nanopillars structure for LED III-LED IV, respectively, compared with LED II; that is, the proposed LEDs with SiO$_2$ nanopillars on microroughened surface show the enhancement in light output power by 42.7–49.1% at 20 mA. The increase of light output power of microrough surface and SiO$_2$ nanopillars structure suggests that the reduction of Fresnel reflection on the surface is a major cause of the increase in light extraction efficiency. It is attributed to more SiO$_2$ nanopillars increasing cause higher light scattering effect by the SiO$_2$ nanopillars on the ITO surface. It also found that the light-extraction efficiency was increased slowly when the height of SiO$_2$ nanopillars increasing due to the occupancy area ratio of SiO$_2$
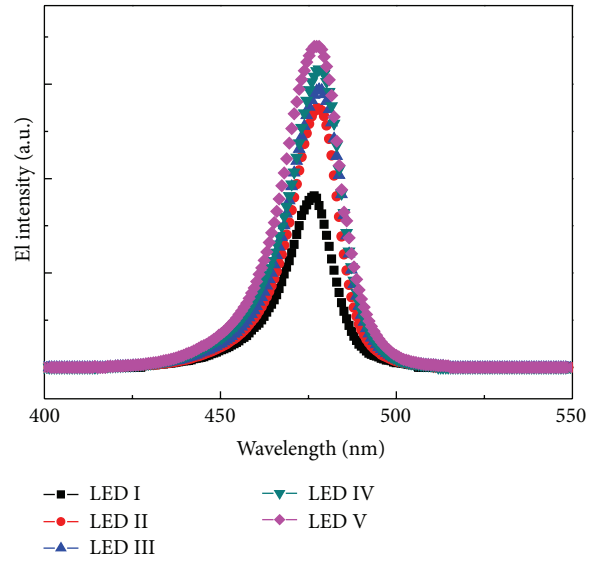
to more roughened surface and optical scattering effects on the proposed GaN-based LEDs.

Acknowledgment

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