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Research Article

Effect of Top-Region Area of Flat-Top Pyramid Patterned Sapphire Substrate on the Optoelectronic Performance of GaN-Based Light-Emitting Diodes

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The flat-top pyramid patterned sapphire substrates (FTP-PSSs) have been prepared for the growth of GaN epilayers and the fabrication of lateral-type light-emitting diodes (LEDs) with an emission wavelength of approximately 470 nm. Three kinds of FTP-PSSs, which were denoted as FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C, respectively, were formed through the sequential wet etching processes. The diameters of circle areas on the top regions of these three FTP-PSSs were 1, 2, and 3 μm, respectively. Based on the X-ray diffraction results, the full-width at half-maximum values of rocking curves at (002) plane for the GaN epilayers grown on conventional sapphire substrate (CSS), FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C were 412, 238, 346, and 357 arcsec, while these values at (102) plane were 593, 327, 352, and 372 arcsec, respectively. The SpeCLED-Ratro simulation results reveal that the LED prepared on FTP-PSS-A has the highest light extraction efficiency than that of the other devices. At an injection current of 350 mA, the output powers of LEDs fabricated on CSS, FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C were 157, 254, 241, and 233 mW, respectively. The results indicate that both the crystal quality of GaN epilayer and the light extraction of LED can be improved via the use of FTP-PSS, especially for the FTP-PSS-A.

1. Introduction

Recently, GaN-based materials have received a lot of attention for many optoelectronic applications consisting of light-emitting diodes (LEDs), laser diodes, and high-power devices [1–3]. Because the price of bulk GaN substrate is very high, the GaN-based epilayers should be prepared on foreign substrates, such as sapphire and silicon carbide. At present, sapphire is the most common substrate for the epitaxial growth of GaN-based LEDs. However, owing to the large mismatch in lattice constant between GaN and sapphire, a large number of threading dislocations (TDs) generated during the epitaxial growth process would lead to a serious degradation of the GaN crystal quality. For the fabrication of GaN-based LEDs, the deterioration of epitaxial quality will obviously limit their optoelectronic performances including internal quantum efficiency (IQE), electron mobility, and device lifetime.

Thus, to reduce the TD density of GaN epitaxial film, several techniques have been presented [4–6]. Among these techniques, the epitaxial lateral overgrowth (ELOG) employing the stripe-mask patterns on the GaN epilayer is a useful method to obstruct the propagation of TDs efficiently, resulting in a significant reduction of TD density [4]. Nevertheless, when the ELOG is performed, the growth interruption caused by the mask deposition is inevitable. Additionally,

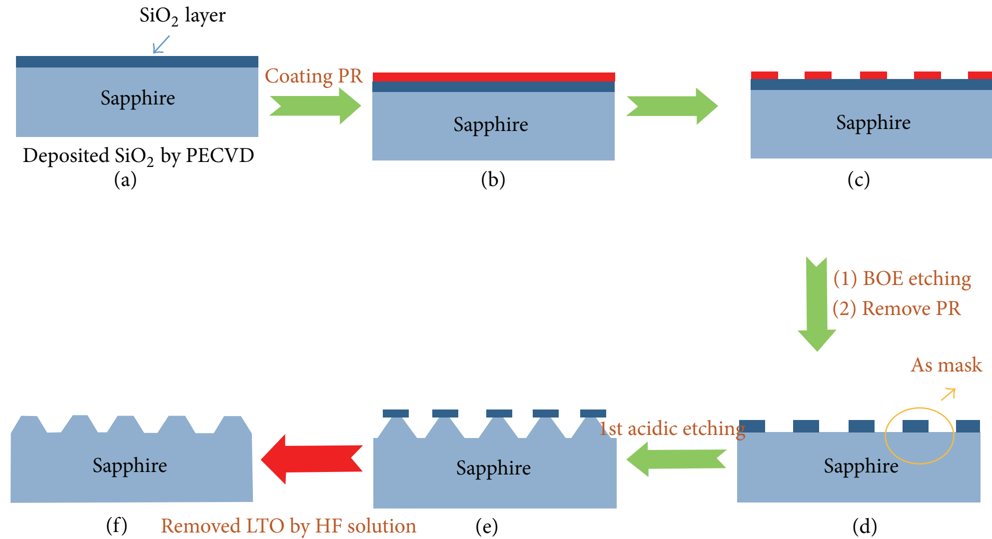

FIGURE 1: Fabrication processes of FTP-PSS: (a) growth of a SiO_2 layer on the *c*-plane sapphire by PECVD, (b) preparation of a thick PR on the SiO_2 layer by spin-coating, (c) performing the photolithography technique to open the PR mask, (d) formation of the round-shaped SiO_2 layer via the buffered oxide etching and removal of the PR mask, (e) wet etching to the sapphire substrate using a mixed $H_2SO_4 : H_3PO_4$ (3 : 1) solution at 250°C for 30 min, and (f) removal of the SiO_2 mask layer using a HF solution.

the formation of mask could also induce the impurity contamination in the subsequent epilayers. To overcome these mask-related drawbacks, the mask-free patterned sapphire substrate (PSS) technology has been proposed and becomes very promising for the growth of high-quality GaN epilayer [7–10]. Except for the improvement in the IQE, the use of PSS also can increase the light extraction efficiency (LEE) of LEDs due to the nonplanar interface between sapphire and GaN.

In previous researches, the InGaN/GaN film with high crystal quality can be achieved using metalorganic chemical vapor deposition (MOCVD) on the flat-top pyramidal PSS (FTP-PSS) [8–10]. These researches confirm that the FTP-PSSs are indeed helpful to enhance the performance of LEDs. However, the influence of pattern size of the FTP-PSS on the device characteristics is seldom studied. In our work, three FTP-PSSs were fabricated to serve as the substrates for the growth of GaN-based epitaxial structures. In these three FTP-PSSs, the heights of patterns are equal to each other. Meanwhile, the diameters of circle areas on both the top and the bottom regions of the flat-top pyramidal patterns in these FTP-PSSs are modified. The effect of the flat-top region area of FTP-PSS on the device performance was investigated.

2. Experimental

The fabrications of PSSs with the flat-top pyramid patterns were completed through the wet etching processes sequentially, as described in Figure 1. As shown in Figure 1(a), the SiO_2 layer with a thickness of 250 nm was firstly prepared on the *c*-plane sapphire by plasma enhanced chemical vapor deposition (PECVD). Then, a thick photoresist (PR) was grown on the SiO_2 layer as a mask by spin-coating (Figure 1(b)). To open the PR mask, the photolithography technique was performed on the PR layer (Figure 1(c)). In Figure 1(d), the round-shaped SiO_2 layer was prepared using a buffered oxide etch (BOE) solution to serve as a mask for the fabrication of PSS, and the PR mask was removed. As shown in Figure 1(e), the sapphire substrate with the SiO_2 mask was etched using a mixed $H_2SO_4 : H_3PO_4$ (3 : 1) solution at 250°C for 30 min. In this research, three different diameters (1, 2, and 3 μm) of round shape were designed for the SiO_2 mask layers. Thus, after removing the SiO_2 mask layer using a HF solution (Figure 1(f)), the FTP-PSSs with three different top-region areas can be formed.

Except for the FTP-PSSs used to fabricate the LEDs, the LED on the conventional sapphire substrate (CSS) was also prepared as the contrasted sample. The LED epitaxial structures grown on FTP-PSSs and CSS were performed by low-pressure MOCVD at the same growth conditions. The LED epitaxial structure consisted of a 3 μm thick undoped GaN (u-GaN) layer, a 2 μm thick n-type GaN:Si layer, 11 periods of InGaN/GaN multiple quantum wells (MQW), a 100 nm thick p-type AlGaN layer, and a 0.2 μm thick p-type GaN:Mg layer. At the beginning of epitaxial growth, it is noted that a 30 nm thick GaN buffer layer was prepared at low temperature (530°C), and then a 3 μm thick u-GaN layer was deposited at high temperature (1150°C). After growing these epilayers, the device processes for lateral-type LED were carried out subsequently. Firstly, a mesa pattern with the size of 45 mil × 45 mil was defined and prepared via the standard photolithography and dry etching processes. Then, the ITO film was grown on the p-type GaN layer to serve as a transparent conducting layer. The ITO film was deposited by E-beam evaporation at 270°C, and its thickness was 200 nm. Finally, the Ti/Al/Ti/Au (15/2000/25/60 nm) metal layers grown on the sample by thermal evaporation were adopted as both n-pad and p-pad electrodes. These LEDs all possess the emission wavelength approximately 470 nm.

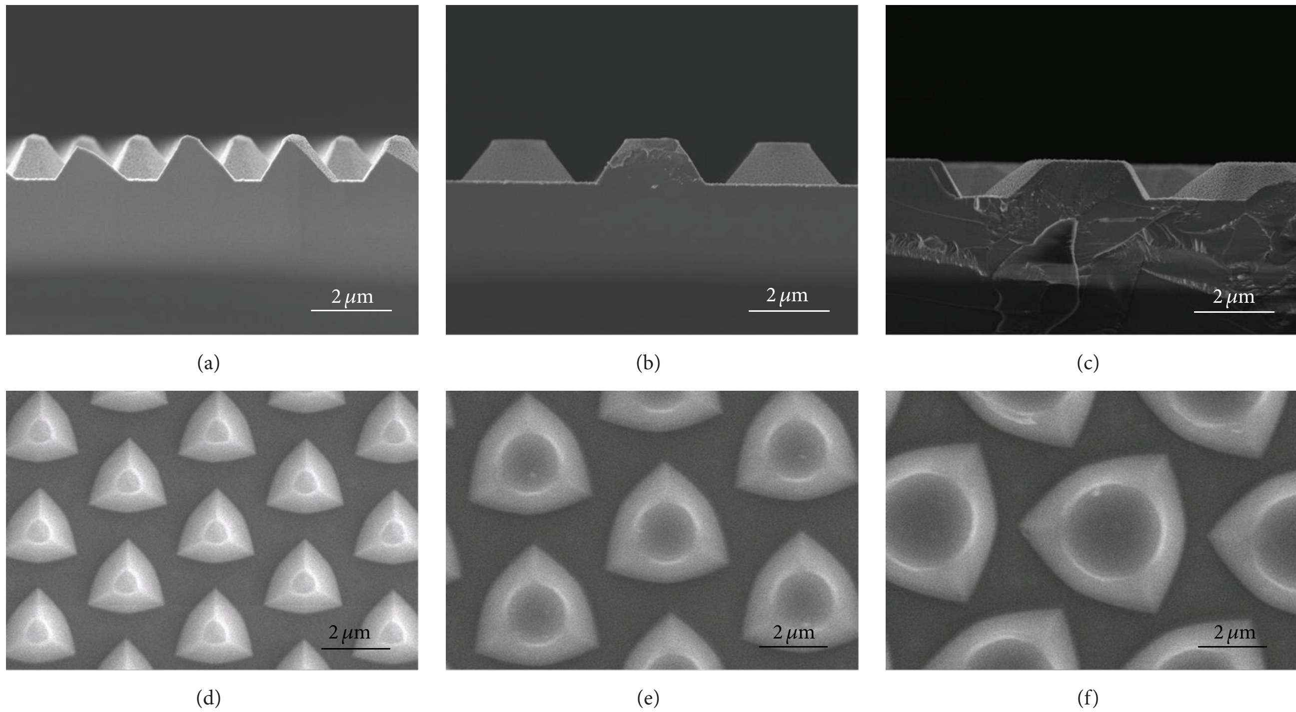

Figure 2: Cross-sectional SEM image of (a) FTP-PSS-A, (b) FTP-PSS-B, and (c) FTP-PSS-C. Top-view SEM images of (d) FTP-PSS-A, (e) FTP-PSS-B, and (f) FTP-PSS-C.

Crystal quality of the u-GaN epilayer was determined by X-ray diffraction (XRD, Philips X'Pert MRD). Here, the analyses of (002) and (102) planes for the u-GaN epilayer were both performed. The surface morphology of the FTP-PSS was observed by scanning electron microscopy (SEM, JEOL JSM-7001F). Current-voltage (I-V) characteristics of these LEDs were investigated by a semiconductor parameter analyzer (Keithley, 2400 SourceMeter), and the output powers were measured with a calibrated integrating sphere. To quantitatively characterize the light extraction efficiency of LEDs on these sapphires, the simulation of photon trajectory was performed by SpeCLED-Ratro software (commercial light-simulation software based on geometrical optics).

3. Results and Discussion

As mentioned above, during the fabrication process of FTP-PSSs, three SiO_2 mask layers with various diameters of 1, 2, and 3 μm of round shape were prepared. Then, three various FTP-PSSs, that were denoted as FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C, respectively, were formed via the sequential wet etching processes. Figures 2(a), 2(b), and 2(c) show the cross-sectional SEM images of FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C, respectively. We can observe that the heights of patterns of these three FTP-PSSs were all fixed at 1.4 μm. Figures 2(d) and 2(e) show the top-view SEM images of these three FTP-PSSs, respectively. The diameters of circle areas on the top and bottom regions of FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C were 1 and 2.4 μm, 2 and 3.8 μm, and 3 and 5.2 μm, respectively. Meanwhile, the distances between patterns of these three FTP-PSSs were 0.85, 0.85, and 0.75 μm, respectively. Obviously, the area of flat-top region for these three FTP-PSSs decreased in the following order: FTP-PSS-C > FTP-PSS-B > FTP-PSS-A. This indicates that the FTP-PSS-C has the largest c-plane area of flat-top region, while the FTP-PSS-A possesses the smallest one among these three samples.

The crystal qualities of the u-GaN epilayers grown on CSS and FTP-PSSs were estimated by measuring the full-width at half-maximum (FWHM) values derived from the XRD rocking curves. The XRD rocking curves obtained for the symmetric GaN(002) plane and the asymmetric GaN(102) plane of these four samples are presented in Figures 3(a) and 3(b), respectively. By growing the u-GaN epilayer on the CSS, the FWHM value at the (002) plane was 412 arcsec, while the FWHM value at the (102) plane was 593 arcsec. Noted that the FWHMs at (002) and (102) of CSS have a little large as compared with those of previously reports [11, 12]. It is well know that the crystal quality of the epilayer is dependent on the MOCVD system. In this study, the growth parameters of CSS have been optimized. On the other hand, the FWHM values at the (002) plane for the u-GaN epilayers grown on FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C were 238, 346, and 357 arcsec, respectively. Additionally, the FWHM values at the (102) plane for the u-GaN epilayers prepared on these three FTP-PSSs were measured to be 327, 352, and 372 arcsec, respectively. It is well known that crystal quality of the (002) plane was influenced by screw and mixed dislocations, while the crystal quality of (102) plane was influenced by edge, screw, and mixed dislocations [13]. It is clear that the crystallinity of u-GaN epilayer grown on the FTP-PSS is much higher than that on the CSS. This can be attributed to the reduction in the dislocation density of GaN epilayer

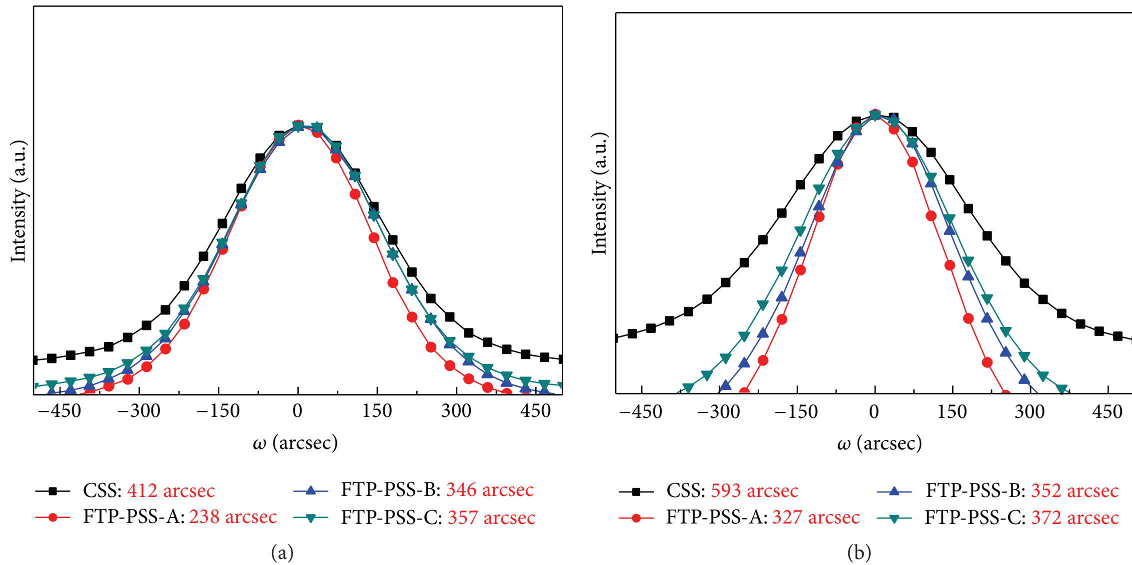

Figure 3: XRD rocking curves obtained for (a) the symmetric GaN(002) plane and (b) the asymmetric GaN(102) plane of GaN epilayers grown on CSS and various FTP-PSSs.

grown on the FTP-PSS. Obviously, the fabrication of FTP-PSS is useful to enhance the crystal quality of GaN epilayer.

Except for the XRD FWHM value, the evaluation of dislocation density also can be used to analyze the crystal quality of u-GaN epilayer. The TD characteristics in u-GaN films on CSS and FTP-PSSs were investigated by etch-pit-density (EPD) measurements. The etching process was performed by soaking the samples into the $H_2SO_4 : H_3PO_4$ (3 : 1) mixed solution at 250°C for 10 min. Figures 4(a), 4(b), 4(c), and 4(d) display the SEM images for the TDs of u-GaN epilayers on CSS, FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C, respectively. It can be found that the EPDs of these four samples were estimated to be 3.25×10^7, 1.39×10^6, 2.31×10^6, and $5.17 \times 10^6\ \mathrm{cm}^{-2}$, respectively. As marked in Figures 4(a) and 4(b), the feature of the screw dislocation is a well-defined hexagonal shape with a large black core, while the mixed dislocation has an unclear hexagonal shape with a small black core. Besides, the edge dislocation shows an unclear polygon [12, 14]. It was obvious that three various types of TDs all can be identified in the GaN epilayer on the CSS (Figure 4(a)). After growing the GaN epilayer on the FTP-PSS, the formation of these three types of TDs reduced efficiently. Moreover, the suppressions of screw and mixed TDs are more obvious than that of edge TDs. Based on XRD and EPD results, the dislocation density of GaN epilayer on the FTP-PSS can be decreased apparently as compared with that on CSS. As the FTP-PSSs were used, the GaN growth could primarily occur in the bottom and flat-top regions (they are both *c*-plane), and the epitaxial lateral overgrowth generated to merge the GaN films [15, 16], resulting in the decrease in the dislocation density.

Figures 5(a)–5(c) display the schematics of initial GaN growth modes on FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C, respectively [17]. As discussed in Figure 2, the FTP-PSS-C possessed a largest pattern size, while the FTP-PSS-A had a smallest pattern size. Moreover, the distances between patterns of these three FTP-PSSs were similar to each other. These reveal that the number of patterns per unit area reduced in the following order: FTP-PSS-A > FTP-PSS-B > FTP-PSS-C. In other words, the area of inclined region in these three PSSs per unit area also decreased in the following order: FTP-PSS-A > FTP-PSS-B > FTP-PSS-C. As described above, a 30 nm thick GaN buffer layer was grown at 530°C, and then the growth temperature was increased to 1150°C. During the heating process, the thin GaN film was gradually transferred to GaN grains. When the subsequent growth was performed at high temperature (1150°C), the GaN epilayer was easily deposited from these GaN grains. Because the FTP-PSS-A possessed the largest area of inclined region (per unit area) compared to that of the other two PSSs, the GaN grains were easily formed in the inclined regions during the heating process. Therefore, the GaN epilayer was easily grown on the inclined regions compared to the bottom *c*-plane regions, as shown in Figure 5(a). However, as shown in Figures 5(b) and 5(c), when the area of inclined region (per unit area) became smaller in FTP-PSS-B and FTP-PSS-C, the initial GaN growth was gradually transferred to GaN on *c*-plane region mode. On the other hand, in the initial growth, the GaN epilayer was also formed on the flat-top region of PSS. Since the FTP-PSS-C had the largest flat-top region area, the GaN film was easily grown on its flat-top regions. On the contrary, the GaN film was more difficultly formed on the flat-top regions of FTP-PSS-A.

The schematics of dislocation behaviors in the GaN epilayers grown on FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C were shown in Figures 5(d)–5(f), respectively [17]. As discussed above, the heights of patterns for these three FTP-PSSs were the same. Meanwhile, the distances between patterns in these three FTP-PSSs were similar to each other. Therefore, the difference in the crystal quality of GaN epilayers grown on FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C could be influenced

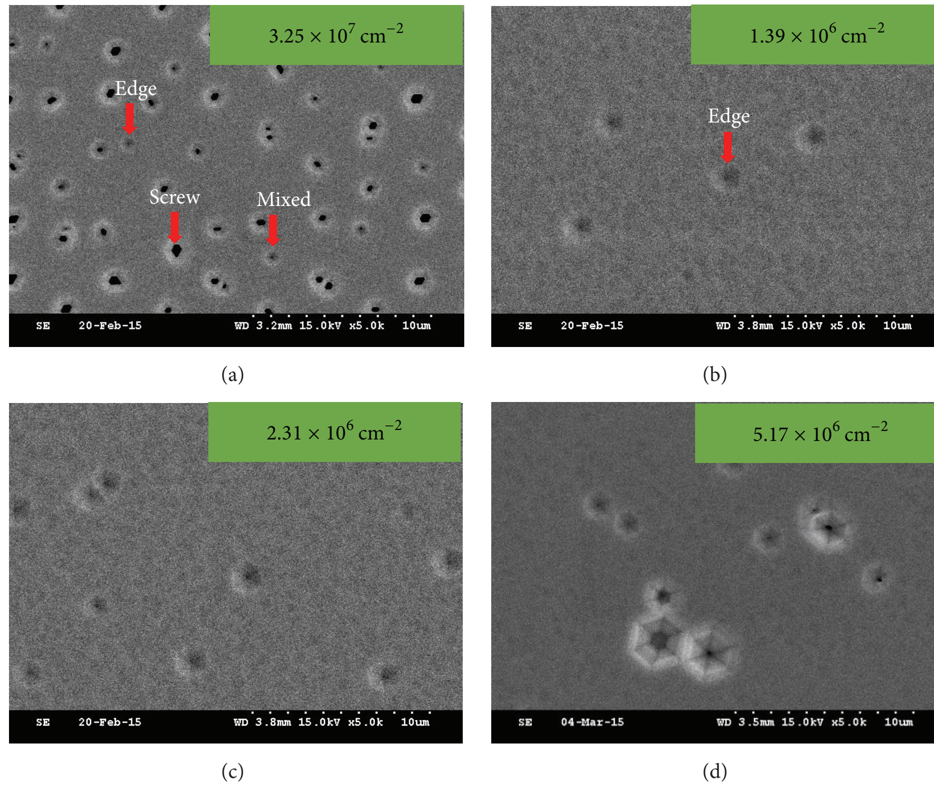

FIGURE 4: SEM images of u-GaN epilayers grown on (a) CSS, (b) FTP-PSS-A, (c) FTP-PSS-B, and (d) FTP-PSS-C after soaking the samples into the $H_2SO_4 : H_3PO_4$ (3 : 1) mixed solution at 250°C for 10 min.

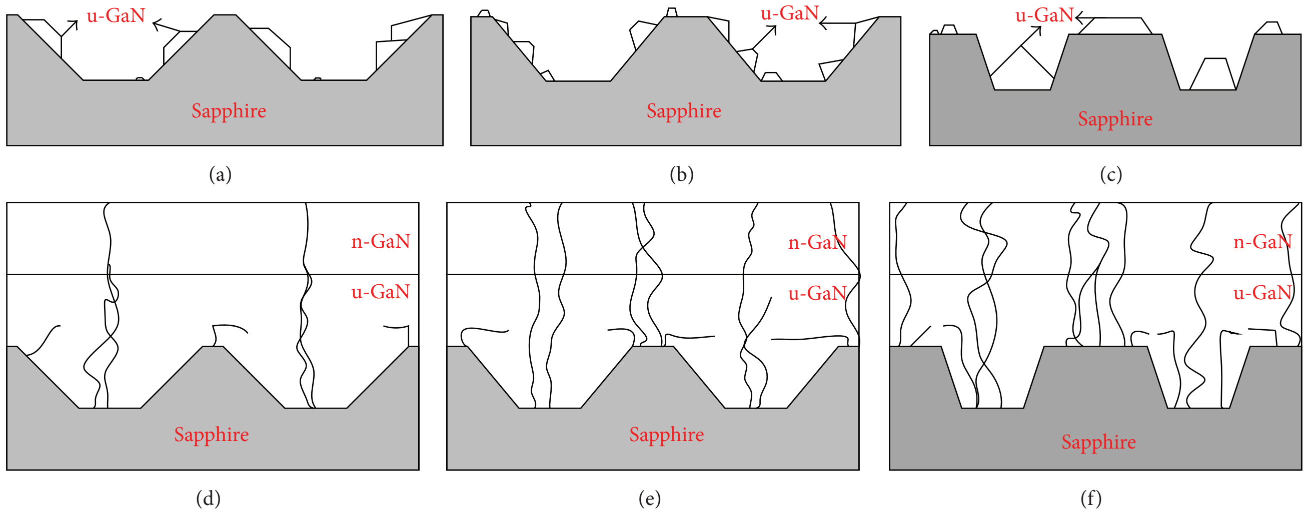

FIGURE 5: Schematic illustrations of the initial GaN growth modes on (a) FTP-PSS-A, (b) FTP-PSS-B, and (c) FTP-PSS-C. Schematic of the evolutions of dislocation behaviors in the GaN epilayers grown on (d) FTP-PSS-A, (e) FTP-PSS-B, and (f) FTP-PSS-C.

by the flat-top region area. Because the FTP-PSS-A possesses the smallest *c*-plane area of flat-top region, the number of dislocations formed at the initial stage is the least, causing the highest crystal quality of GaN epilayer grown on this PSS. On the contrary, due to the largest *c*-plane area of flat-top region formed in the FTP-PSS-C, the GaN epilayer prepared on this PSS has a relatively low crystal quality as compared with that on the other PSSs.

Figure 6 shows the SpeCLED-Ratro simulation results for the LEDs. During the simulation, a 350 mW power was used to emit randomly from the MQW region. As shown in Figure 6(a), the simulated output power and LEE of the LED

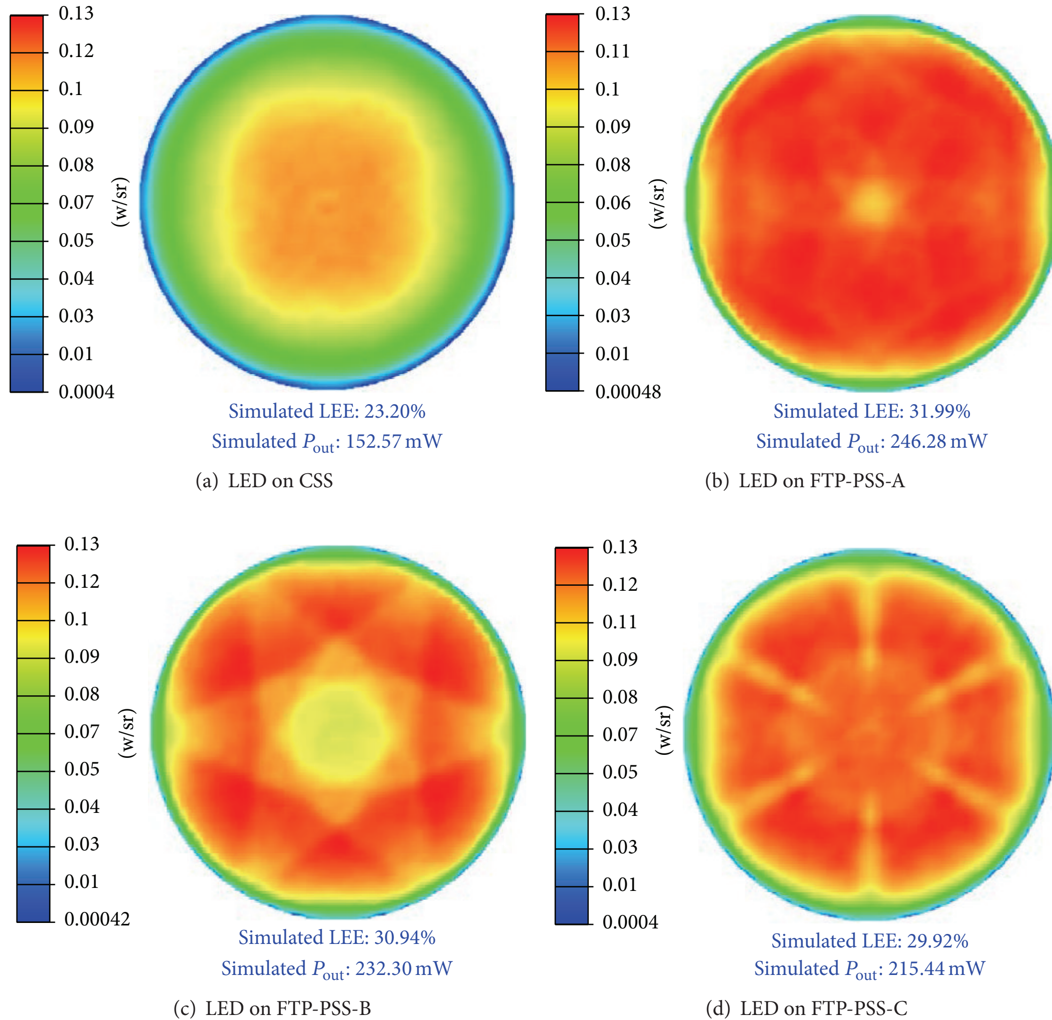

Figure 6: SpeCLED-Ratro simulation results for the LEDs fabricated on (a) CSS, (b) FTP-PSS-A, (c) FTP-PSS-B, and (d) FTP-PSS-C.

on CSS were 153 mW and 23.2%, respectively. The simulation results for the LEDs prepared on various FTP-PSSs are shown in Figures 6(b)–6(d). We can observe that the simulated output powers of LEDs on FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C were increased to 246, 232, and 215 mW; meanwhile, the simulated LEEs of these LEDs were enhanced to 32.0%, 30.9%, and 29.9%, respectively. The results present that the LED prepared on the FTP-PSS-A possesses the highest LEE than that of the other samples.

Figure 7 displays the *I-V* characteristics of the LEDs fabricated on CSS, FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C. Although the forward voltages of these four devices measured at the whole injection currents (0–1050 mA) are similar to each other, there is a slight difference in the forward voltages of LEDs fabricated on CSS and FTP-PSSs. At an injection current of 350 mA, the forward voltages of the LEDs prepared on the CSS, FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C are measured to be 3.27, 3.22, 3.25, and 3.27 V, respectively. Further increasing the injection current to 1050 mA, the forward voltages of these four devices were 4.30, 4.19, 4.26, and 4.28 V, respectively. This indicates that the crystal quality of GaN epilayer also affects the *I-V* characteristic of LED. Apparently, the higher the crystal quality is obtained in the GaN epilayer, the lower the forward voltage can be achieved in the LED.

Figure 8 shows the light output powers as a function of injection current for these four LED samples fabricated on the CSS and various FTP-PSSs. The light emission images (at 20 mA) of these four LEDs are shown in the inset. Obviously, the light emission intensity of LED on FTP-PSS is much higher than that on CSS. The output powers (at 350 mA) of LEDs prepared on CSS, FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C were 157, 254, 241, and 233 mW, respectively. In comparison to the LED on CSS, there are 61.8%, 53.5%, and 48.4% enhancements in the output power (at 350 mA) for the devices on FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C, respectively. As the injection current was increased to 1050 mA, the output powers of these four LEDs were 371, 581, 543, and 505 mW, respectively. Based on the above-mentioned results, the use of FTP-PSS can both enhance the

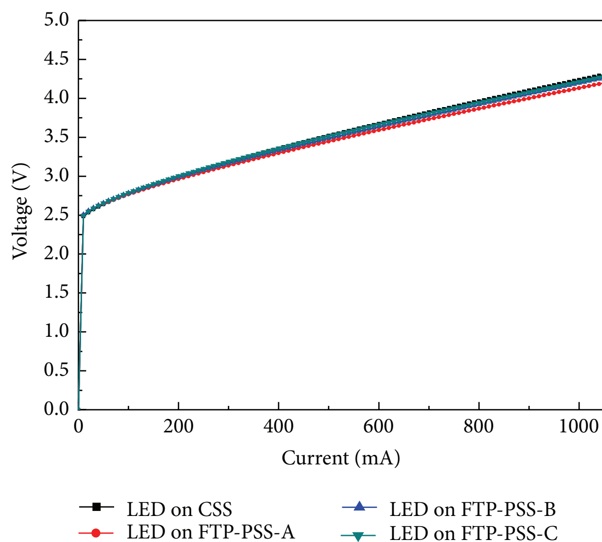

FIGURE 7: I-V characteristics of the LEDs fabricated on CSS and various FTP-PSSs.

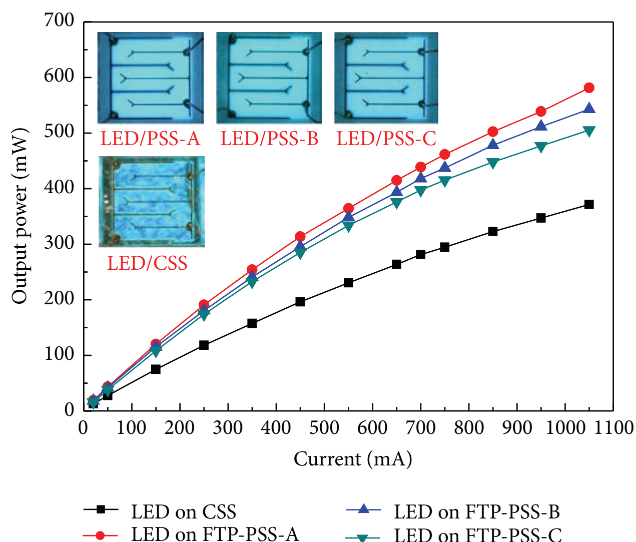

FIGURE 8: Light output powers as a function of injection current for the LEDs fabricated on CSS and various FTP-PSSs.

epilayer quality and increase the light extraction of LED. In particular, for the LED fabricated on FTP-PSS-A, it reveals that this device has better optoelectronic performances.

4. Conclusion

In summary, by performing the sequential wet etching processes, the FTP-PSSs have been prepared for the fabrication of GaN-based blue LEDs. The influence of the flat-top region area of FTP-PSS on the LED performance was analyzed. After fabricating the LEDs on FTP-PSSs, the optoelectronic performance of device was enhanced remarkably as compared with that prepared on CSS. Particularly, for the use of FTP-PSS-A, the crystal quality of GaN epilayer and the light extraction in the LED were both improved most obviously. At an injection current of 350 mA, the output power of LEDs prepared on CSS was measured to be 157 mW. Additionally, as the LEDs fabricated on FTP-PSS-A, FTP-PSS-B, and FTP-PSS-C, the output powers (at 350 mA) were increased to 254, 241, and 233 mW, respectively. Compared with the LED on CSS, there is 61.8% increment in the output power (at 350 mA) for the devices on FTP-PSS-A.

Competing Interests

The authors declare that they have no competing interests.

Acknowledgments

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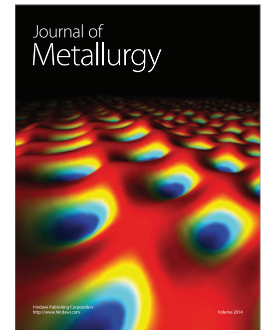
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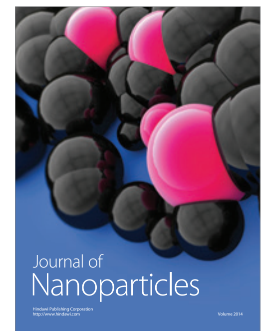
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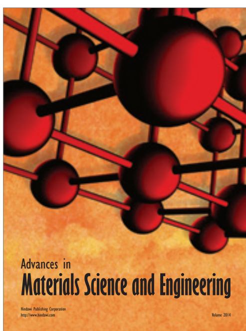
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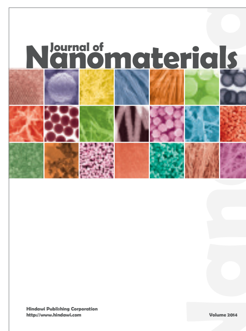
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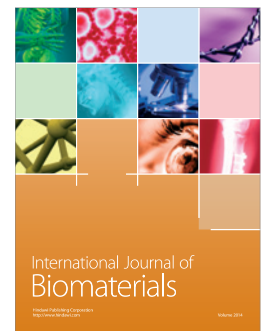
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