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Research Article

SILAR-Based Application of Various Nanopillars on GaN-Based LED to Enhance Light-Extraction Efficiency

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We reported the various nanopillars on GaN-based LED to enhance light-extraction efficiency prepared by successive ionic layer adsorption and reaction method (SILAR). Indium tin oxide (ITO) with thickness of 1 μm as transparent contact layer was grown to improve the electrical characteristics of the LEDs, including series resistance and operating voltage. SILAR-deposition ZnO nanoparticles on SiO_2 were used as etching nanomasks. Multiple nanopillars were simultaneously formed on overall surfaces of ITO p- and n-GaN by ICP etching. The proposed GaN-based LEDs with nanopillars increase light output power by 7%–20.3% (at 20 mA) over that of regular GaN-based LEDs. The difference in light output power can be attributed to differences in materials and shapes of nanopillars, resulting in a reduction in Fresnel reflection by the roughened surface of GaN-based LEDs.

1. Introduction

GaN-based materials and their 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]. It is required to further extend the application arm of GaN-based LED to projectors automobile headlight and even general lighting and further improvement on optical power and light-extraction efficiency are eagerly required. Several methods, including surface roughening techniques [2–5], inclined sidewall etching [6], patterned sapphire substrates [7, 8], and the incorporation of highly reflective omnidirectional reflectors (ODRs) [9], have been shown to effectively improve the light-extraction efficiency. Among these methods, surface roughening is one of the most efficient to provide an enhancement factor for the extraction efficiency due to increased random scattering events that occur at the roughened surfaces. However, the etching techniques have inherent limit to the thickness of p-GaN layer, which is ~300 nm [10–12]. Therefore, the etching process must be precisely controlled to avoid deterioration of electrical properties. Moreover, although other approaches involving the synthesis of ZnO nanorods and the spin coating of polystyrene nanospheres have been developed for GaN-based LEDs, separate process steps and foreign materials are unavoidable [13–15]. On the other hand, indium tin oxide (ITO) has been widely used in GaN-based LEDs as transparent conductive layer (TCL) to improve current spreading in p-GaN layer. Popular-use thickness of ITO on p-GaN is about 200–245 nm. However, the thickness is too thin to etch due to influence of the current spreading of GaN-based LEDs. In this paper, we proposed the highly thick ITO as the TCL of GaN LEDs. Because both LED operation voltage and series resistance could be reduced due to the reduction in current crowding, on the other hand, the thickness provides the enough depth for the roughening surface on ITO without any damages in electrical properties. In order to produce nanoscale patterns on the surface of GaN-based LEDs, so far, few researches report chemical and physical deposition techniques on the fabricating of ZnO nanoparticles. As our previous report [16], SILAR-deposition ZnO nanoparticles were used to produce the SiN_x nanopillars on the surface of GaN-based LEDs. The SILAR method approaches based on the soft chemical technique have attracted increasing attention [17], due to its high reliability, low cost, and large-area deposition compared with other methods. In this paper, high-thickness ITO was used as the transparent contact layer of GaN LEDs, and different densities of self-assembly ZnO nanomasks and etching time were used to produce the roughened surface including the ITO surface, p-GaN layer around the edge of the mesa, the sidewall, and the n-GaN layer on GaN-based LEDs. In addition, the resulting light output power efficiency of LEDs with roughness surfaces is significantly higher than that of a conventional LED.

2. Experimental

The GaN-based LED was grown on c-axis sapphire substrates by using low-pressure metal-organic chemical-vapor deposition. N-type GaN epitaxial layers, which included a 1 μm thick undoped GaN layer and a 2 μm 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 and a 150 nm p-GaN layer were fabricated. The LEDs had a mesa structure with an area of $300 \times 300\,\mu\text{m}^2$. Before fabricating the LEDs using SiO_2, a 1 μm thick ITO film was deposited on the top of GaN-based LEDs by radio frequency magnetron sputtering, and we also prepared a 200 nm thickness of ITO film LED to be compared. Cr/Au layers as the p- and n-contact electrodes were fabricated. Figure 2 demonstrates the schematic diagram of the pattern transfer procedure. A 300 nm thick SiO_2 film was deposited on an ITO layer by PECVD as Figure 2(b). The metal contacts were covered with a photoresist layer by photolithography in order to prevent the SiO_2 which was grown on the ohmic contact electrodes. In Figure 2(c), the ZnO nanoparticles were grown on the top of SiO_2 film as the etching mask by SILAR. The detailed procedures of ZnO nanoparticles in one cycle are shown in Figure 1 and described in our previous report [16]. In this case, the samples were dipping in 95°C DI water : ethylene glycol = 1 : 1 for 20 s to form ZnO nanoparticles on SiO_2 film. Furthermore, we etched SiO_2 by using ICP to get the SiO_2 nanopillars on all the surfaces using a CF_4 gas in the first case as shown in Figure 2(e). We removed the ZnO nanoparticles and photoresist by HCL and acetone. As shown in Figure 2(f), the second case, after etching SiO_2, we intentionally fabricated nanostructures on both ITO and GaN simultaneously using a Cl_2–Ar gas mixture. In the third case, after the second case, we removed the SiO_2 nanoparticles and photoresist by BOE and acetone as shown in Figure 2(g). 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 four cases of the 30° tile-view SEM images of the surface on GaN-based LEDs at ratio of DI water : ethylene glycol = 1 : 1; (a) conventional LED I (with 240 nm thick ITO) and LED II (with 1000 nm thick ITO); (b) LED III (with 1000 nm thick ITO) with SiO_2 nanopillars; (c) LED IV (with 1000 nm thick ITO) with SiO_2/ITO, SiO_2/p-GaN, and SiO_2/n-GaN nanopillars; (d) LED V (with 1000 nm thick ITO) with ITO, p-GaN, and n-GaN by removing SiO_2. Figure 3(a) shows the first case, LED I and LED II, and Figures 3(a1) and 3(a2) display the magnified images of the flat surface ITO, p-GaN, and n-GaN regions, respectively. After the ZnO nanoparticles were deposited at 95°C DI water : ethylene glycol = 1 : 1 and etching, Figure 3(b) (LED II) shows that the diameter and height of SiO_2 nanopillars were from 100 to 450 nm and 300 nm, respectively, and the spacing was approximately 1 μm. Figures 3(b1) and 3(b2) show the magnified images of SiO_2 nanopillars on the surfaces of ITO, p-GaN, and n-GaN, respectively. Subsequently, we intentionally fabricated nanostructures on both ITO and GaN simultaneously using a Cl_2-Ar gas mixture and Figure 3(c) (LED III) shows that diameter and height of nanopillars were from 100 to 350 nm and 250 nm, respectively. The spacing was also approximately 800 μm. Figures 3(c1) and 3(c2) display the magnified images of SiO_2/ITO, SiO_2/p-GaN, and SiO_2/n-GaN nanopillars on surface, respectively. Furthermore, we removed the SiO_2 by BOE from top of surface of GaN-based LED which was shown in Figure 3(d) (LED IV). Figures 3(c1) and 3(c2) display the magnified images of the ITO, p-GaN, and n-GaN nanopillars on surface, respectively. The average depth of the ITO pattern is around 200 nm. We can figure out from Figures 3(c1) and 3(d1) that after removing SiO_2, the shape of patterns would become ladder-shaped and we will analyze the effect of this situation in detail later.

Figure 4(a) shows the current-voltage (I-V) characteristics of thinness of conventional LED and thickness of ITO of conventional LED with and without nanostructures. At a current of 20 mA injection, it was found that forward voltages were 3.5, 3.22, 3.28, 3.27, and 3.27 V for LED I, LED II, LED III, LED IV, and LED V, respectively. It showed that both of the forward voltages and series resistance of thickness of ITO of conventional LED are lower than thinness of conventional LED because much more effectiveness for the current crowding reduction is increasing the electron concentration in the contact layer of the LED, and it also found that the I-V characteristics of GaN-based LEDs with and without nanopillars were very similar. The slightly higher forward voltages could result from that lone time dry etching damage to ITO film, which affects the sheet resistance [18] and current spreading and even GaN film. Figure 4(b) demonstrates light output power of thinness of conventional LED and thickness of ITO of conventional LED with and without nanopillars at 20 mA driving current. The output intensities of these LEDs increased with injection current when the injection current was small and can be seen. Furthermore, it was found that output powers observed from the three cases of LEDs with 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 6.61, 6.48, 6.96, 7.73, and 7.12 mW for the LED I, LED II, LED III, LED IV, and LED V, respectively. It showed that light output power of thickness of ITO of conventional LED is lower than thinness of conventional LED about 2% due to free carrier absorption of emitted light in the ITO film. It was also shown that smaller output power of LED I and LED II is attributed to Fresnel

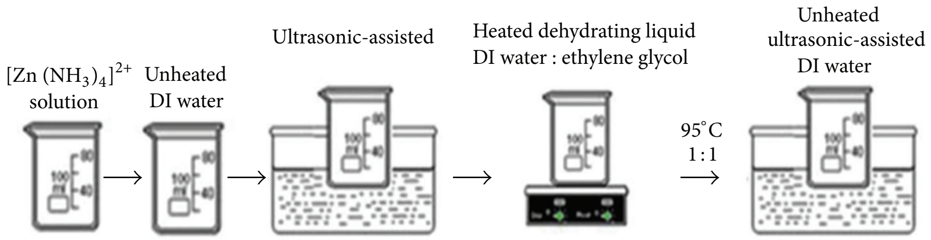


FIGURE 1: Process schemes of the various rinsing procedures.

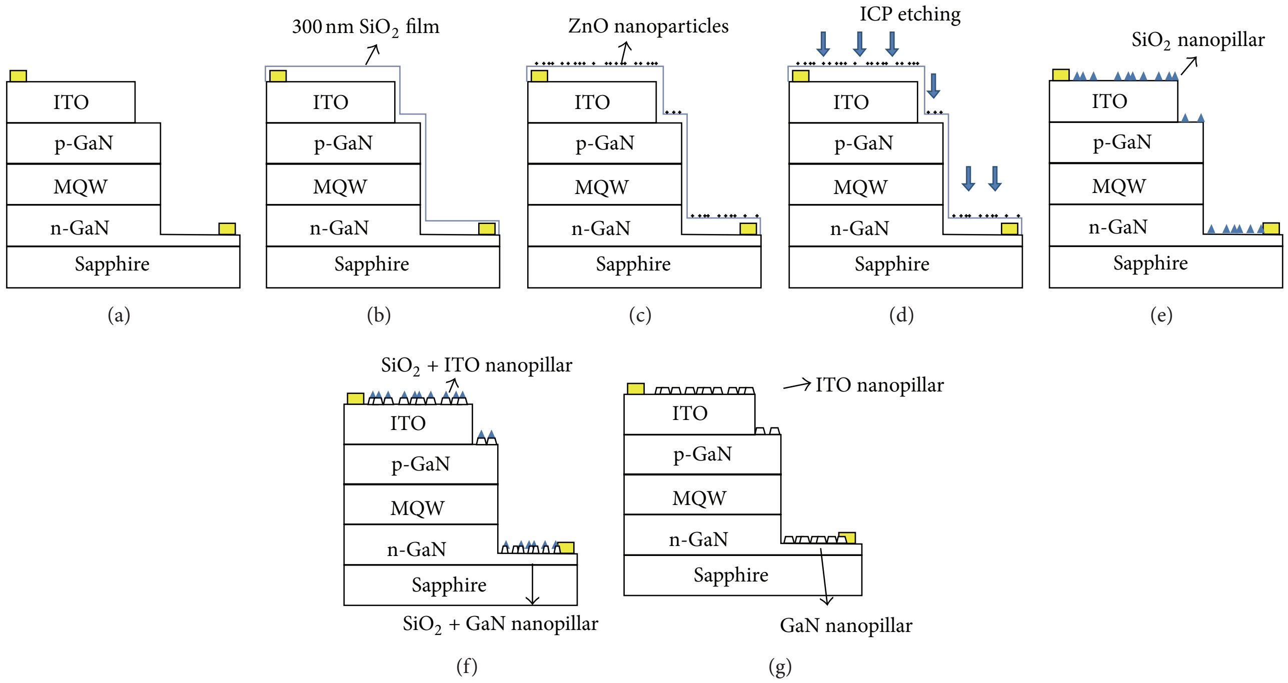
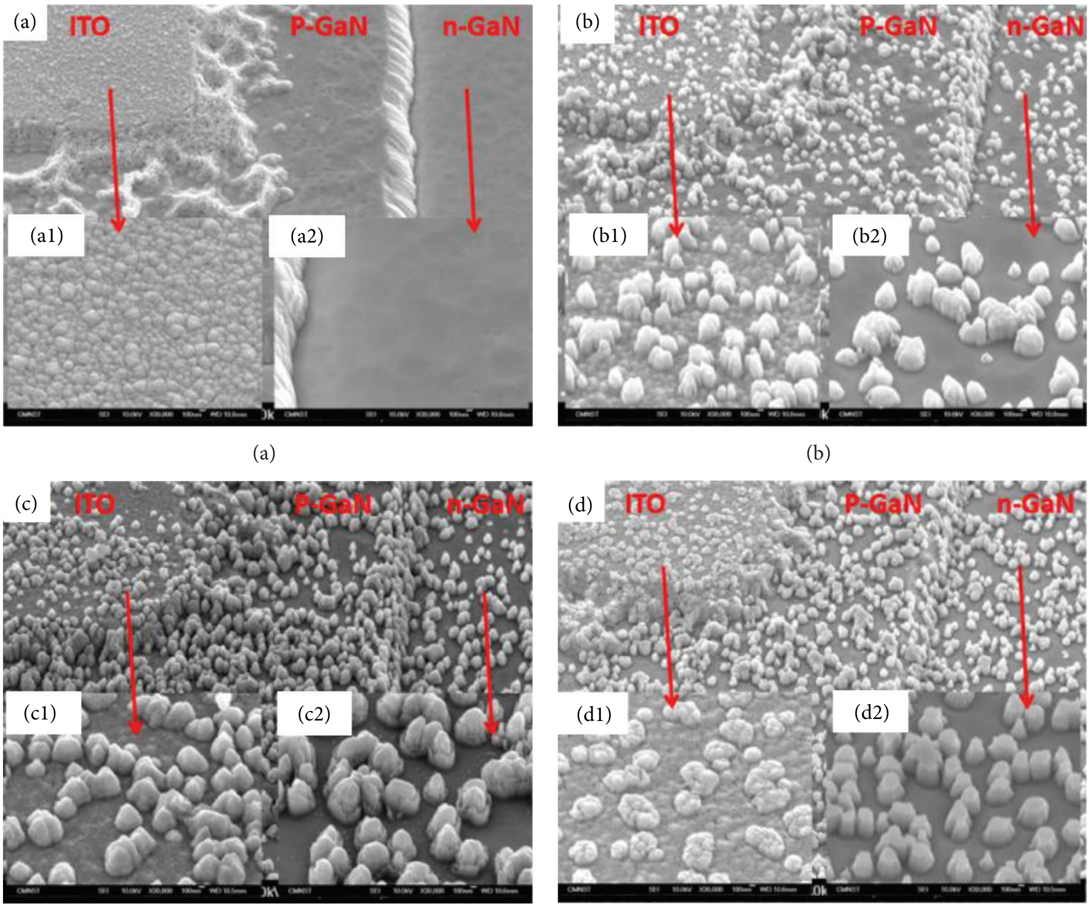


FIGURE 2: Schematic diagram of the pattern transfer procedure of nanostructure LED: (a) conventional LED (LED-II); (b) deposited 300 nm SiO$_2$ on the surface by PECVD; (c) deposition of ZnO nanoparticles on the surface by SILAR; (d) ICP etching; (e) LED with SiO$_2$ nanopillars (LED-III); (f) LED with SiO$_2$, ITO, and GaN nanopillars (LED-IV); (g) LED with ITO and GaN nanostructure (LED-V).

reflection. In other words, we can enhance the light output power at 20 mA by 7%, 20.3%, and 9.1% with nanostructures for LED III–LED V, respectively, compared with LED II. The increase of light output power of nanostructure suggests that the reduction of Fresnel reflection on the surface is a major cause of an increased light-extraction efficiency. It also found that the light-extraction efficiency was increased following the higher density of nanostructure. It is attributed to more nanopillars increasing cause of higher light scattering effect by the nanopillars on the ITO and GaN surfaces.

Figure 5 shows the schematic cross-section diagrams of the LEDs in conventional LED and LED with nanopillars, respectively. As Figure 5(a), most of the light generated in the MQWs active layers is trapped inside the device because of the refractive index difference between the semiconductor and the surrounding medium. For nano tructures LEDs, the surfaces of ITO, p-GaN, and n-GaN were all nanopatterned. The trapped guided modes could multiply scattering and find more chances for radiation out from the device and they suggest that the reduction of Fresnel reflection on the surface is a major cause of increased light-extraction efficiency.

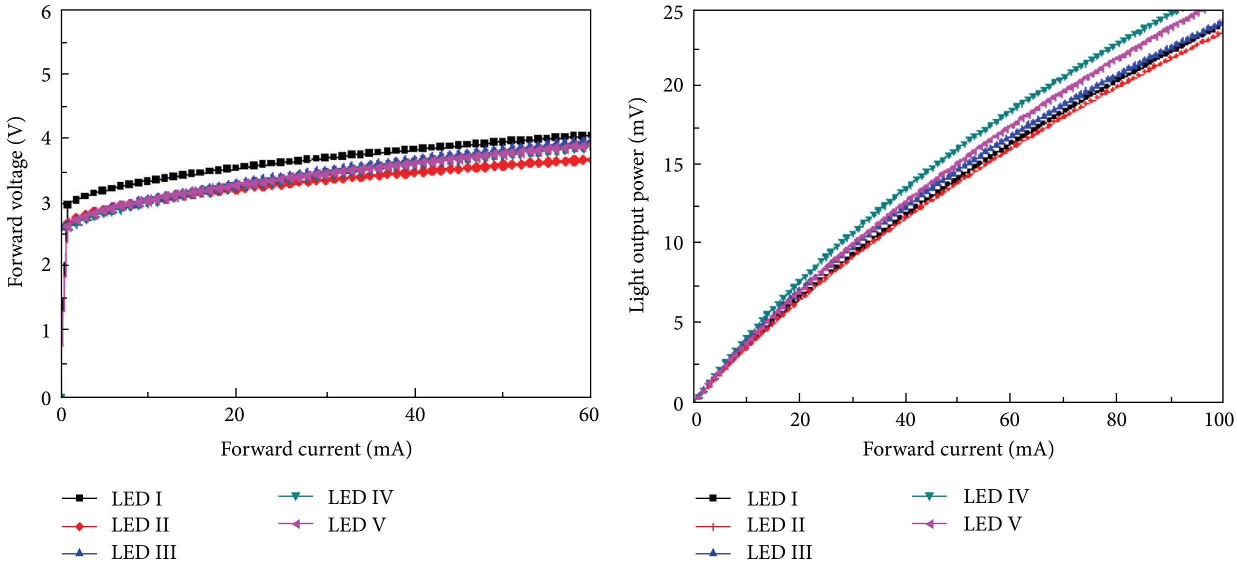
Figure 6 shows the schematic cross-section diagrams of the LEDs. In Figure 6(a), it shows that most of the light generated in the MQWs active layer is trapped inside the device due to the refractive index difference between the semiconductor and the surrounding medium. The SiO$_2$ nanopillars are useful for light extraction, since more surfaces of the nanopillars can provide more opportunities for light escaping and reduce Fresnel refraction because SiO$_2$ refractive index was lower than ITO as shown in Figure 6(b), but partial plane ITO of surface can cause Fresnel refraction. In Figure 6(c), it shows more enhancing of light output power by further etching of ITO because ITO became rough resulting in reducing Fresnel refraction. Furthermore, we removed the SiO$_2$ nanopillars by BOE and got the ladder-shaped ITO patterned. It was found that light output power reduced because the surface of the ladder-shaped ITO was plane and caused the Fresnel reflection again.



(a) (b)

(c) (d)

FIGURE 3: SEM images of the surface: (a) flat surface: LED I and LED II without nanopatterns, (b) LED III with SiO_2 nanopillars, (c) LED IV with SiO_2/ITO, SiO_2/p-GaN, and SiO_2/n-GaN nanopillars, and (d) LED V with ITO, p-GaN, and n-GaN nanopillars by removing SiO_2.



(a) (b)

FIGURE 4: (a) *I-V* curves of GaN-based LEDs with and without nanopillar SiO_2. (b) Light output current characteristics of GaN-based LEDs with and without nanopillars.

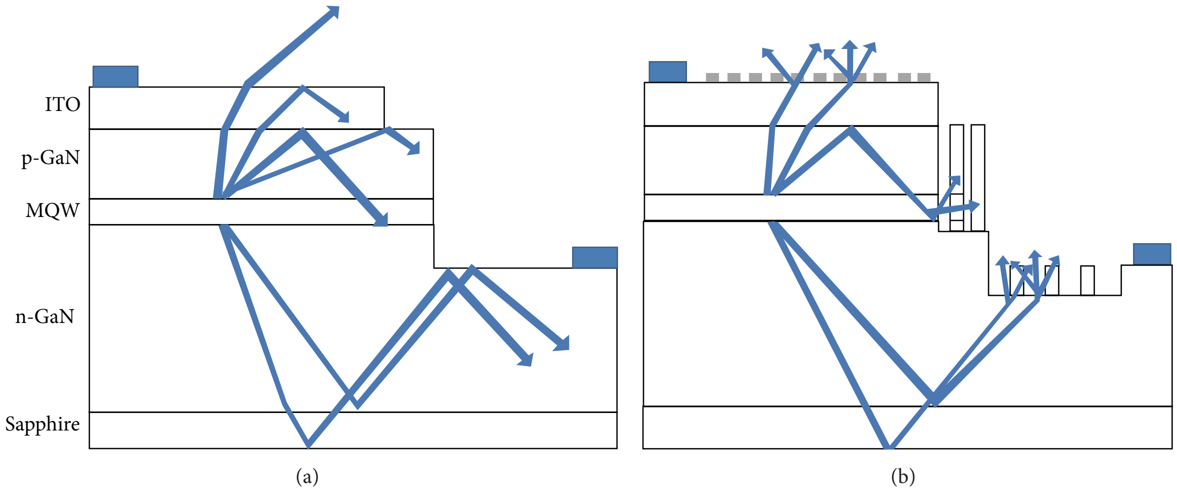


Figure 5: Schematic diagrams of (a) conventional GaN-based LED and (b) GaN-based LED with nanopillars.

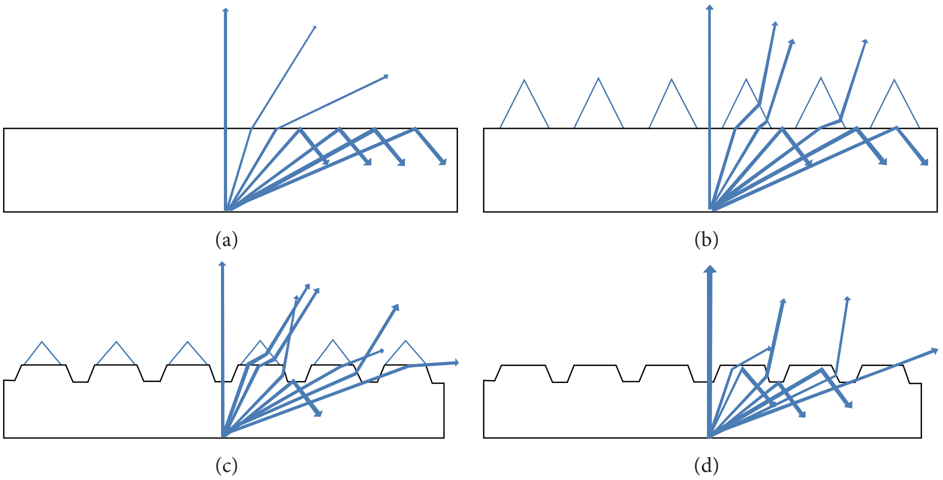


Figure 6: Schematic diagrams of the difference of material and shape of nanostructures: (a) LED with flat surface ITO, (b) LED with SiO_2 nanopillars, (c) LED with SiO_2/ITO nanopillars, and (d) LED with ITO nanopillars.

4. Conclusion

In summary, it was found that thick ITO as TCL can reduce current crowding and series resistance. By using ZnO nanoparticles as dry etching nanomasks on SiO_2 film before SiO_2 etching by ICP, and then further etching ITO and GaN. In addition, we have successfully demonstrated a feasible method to enhance light extraction by producing nanostructures on overall GaN-based LED surfaces by low-temperature SILAR method. The optimal enhancement of light output power of the GaN-based LEDs with nanostructures is achieved up to 20.3% compared to that of the conditional LED at injection current of 20 mA. The enhancement is attributed to the reduction of Fresnel reflection and scattering effects by nanopillars on the whole surface of the LED.

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