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Light extraction improvement of InGaN light-emitting diodes with large-area highly ordered ITO nanobowls photonic crystal via self-assembled nanosphere lithography

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The InGaN multiple quantum well light-emitting diodes (LEDs) with different sizes of indium-tin-oxide (ITO) nanobowl photonic crystal (PhC) structure has been fabricated using self-assembled monolayer nanosphere lithography. The light output power (LOP) of PhC LEDs (at 350 mA) has been enhanced by 63.5% and the emission divergence exhibits a 28.8° reduction compared to conventional LEDs without PhC structure. Current-Voltage curves have shown that these PhC structures on ITO layer will not degrade the LED electrical properties. The finite-difference time-domain simulation (FDTD) has also been performed for light extraction and emission characteristics, which is consistent with the experimental results. © *2013 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License.* [http://dx.doi.org/10.1063/1.4823478]

I. INTRODUCTION

The high brightness GaN light-emitting diode now has been widely used in solid-state lighting, full color displays, and backlights for liquid crystal displays.[1] In order to achieve greater energy efficiency than conventional fluorescent lighting lamps, the GaN-based white LEDs needs further improvement in external efficiency (EQE), which is determined by both internal quantum efficiency (IQE) and light extraction efficiency (LEE). By optimizing the epitaxial layer structure and improvement of crystal quality, LEDs on sapphire substrates whose IQE has reached above 70% have been reported.[2] However, EQE is still relatively low due to the total internal reflection at the interface between GaN layer, indium tin oxide (ITO) layer and the air. At the GaN ($n_{GaN} = 2.5$)/ITO ($n_{ITO} = 1.8$) interface, the critical angle determined by Snell's law [$\theta_c = \sin^{-1} n_{ITO}/ n_{GaN}$] is 70.1°.[3] Correspondingly, the critical angle of ITO and air ($n_{air} = 1$) is 31.8°. Therefore, there is much room for improving the LEE at the interface of ITO layer and the air. It has been demonstrated that introducing a 2D photonic crystal (PhC) structure in the ITO layer is an effective way to improve the LEE of LEDs by coupling the guided modes into radiation modes. Numerous approaches have been adopted to fabricate ITO PhC structures[4,5] such as electron beam lithography (EBL), nano imprint lithography, laser processing technique and laser holography lithography process.[6–13] However, the methods mentioned above either require high price or complicated preocess, leading to a higher fabrication

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cost of LED chips. Compared to above methods, nanosphere lithography (NSL) we introduce in this study is a suitable and economic way to fabricate PhC structures in ITO layer with advantages of low-cost, high-throughput, and large-scale coverage for commercialization.[14,15] In this letter, we have fabricated wafer-scale ITO nanobowls PhC structures in GaN-based LEDs using NSL process. Furthermore, our work has also optimized the nanobowl PhC structures on the ITO layer. An enhancement of 63.5% in the light extraction from LEDs with highly ordered nanobowls ITO PhCs is demonstrated, through electroluminescence and light output power-current-voltage measurements. Meanwhile, simulations based on finite-difference time-domain (FDTD) methods are adopted to investigate the effect of incorporating ITO PhCs of different dimensions on the characteristic of LEDs.

II. EXPERIMENTAL

The samples were grown on c-plane (0001) sapphire substrates by MOCVD. The epitaxial LED structure is similar to our previous report.[16] After a 510 nm-thick ITO transparent conductive layer was deposited on the wafer, ITO nanobowls PhCs were then fabricated using nanosphere lithography-based secondary graphics transfer, as shown in Fig. 1(a)–1(d). Firstly, wafer-scale hexagonal closed-packed monolayer of polystyrene (PS) spheres with a diameter of 1 μm were self-assembled at the gas/liquid interface and be transferred onto the ITO surface. After being dried in the air, the PS microsphere monolayer was obtained. For the preparation of inverted self-assembled monolayer template, SiO_2 sol diluted by isopropanol was coated on the PS microsphere monolayer with a speed of 6000 r/min for 30s. By annealing in air at 600 °C for 30 min for solidification of SiO_2 sol, the PS nanospheres were completely removed, and retained SiO_2 nanobowl which directly served as an etching mask for pattern transfer onto the ITO layer by inductively coupled plasma (ICP) etching. The ICP etching process was performed using a gas mixture of BCl_3 (25 sccm) and Cl_2 (15 sccm) at an RF power of 50 W, for durations of 400s, 800s, and 1200s, respectively. To remove SiO_2 residual to avoid degrading p-electrode ohmic contact, the wafer was soaked in BOE for 40s. Therefore, after the SiO_2 residual was removed, a large wafer-level array of nanobowls was fabricated on the ITO transparent conductive layer. The LED wafers were then processed into chips with a square mesa of 45mil×45mil in size and n-GaN layer was exposed by photolithography and inductively coupled plasma reactive ion etching (ICP). Metal contacts composed of Cr/Pt/Au (50/50/1500 nm) were evaporated onto the ITO transparent conductive layer and the n-type GaN layer by an e-beam evaporator. For comparison, un-patterned LEDs with the identical dimension were also fabricated as reference samples.

III. RESULT AND DISCUSSIONS

Figure 2(a) shows SiO_2 nanobowls on a whole two-inch epitaxial wafer acted as an etch mask for transferring pattern onto the ITO layer. In order to get fewer defects in the SiO_2 nanobowls array, hexagonal monolayer of PS spheres should minimize the occurrence of defects such as dislocations and vacancies and multiple-layer PS spheres should be avoided. Therefore, when transferred monolayer PS spheres onto the LED wafer with ITO layer, the pulling motion must be at a uniform velocity optimized at 1cm/min. As shown in Fig. 2(b), ITO nanobowls are achieved after the ICP etch and the SiO_2 removal process. The inset shows the SiO_2 nanobowls mask on the LED wafer before the ICP etching process.

The influence of the ITO nanobowl surface morphology depends strongly on the ICP power, a gas mixture of Cl_2, BCl_3 and Ar_2 gas flow rate, chamber pressure ICP power and bias voltage, and the etching time. The higher ICP power would make the pattern process less controllable. However, using a lower ICP power would lead to longer etching time. In our study, patterns were transferred onto ITO layer by ICP dry etching with gases in combination with Ar (5 sccm), BCl_3 (5 sccm), and Cl_2 (40 sccm) at a pressure of 4mTorr, with a source power of 50 W and platen power of 300 W. Figure 3(a)–3(d) show the surface morphology of the planar ITO and ITO layers etched for 400s, 800s, and 1200s, respectively, by atomic force microscope(AFM). The inset image depicts the depth of nanobowls for 400s. The depth of the ITO nanobowls can be estimated from the 3D AFM images

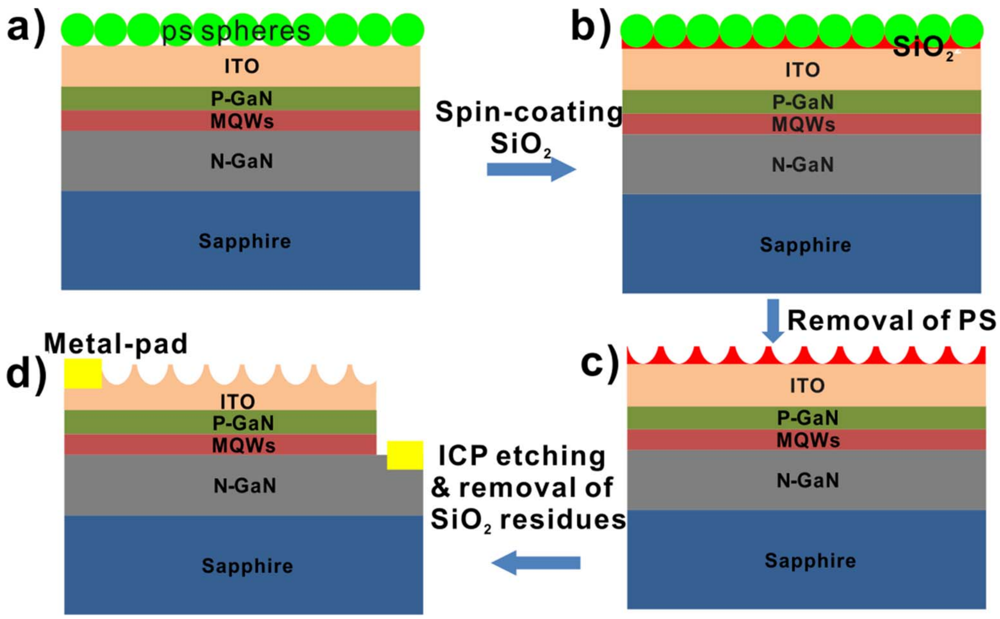

FIG. 1. Schematic diagrams of the fabrication process flow: (a) PS nanospheres self-assembly onto the ITO layer, (b) spin-coated SiO_2 sol, (c) removal of PS nanospheres, and (d) ICP etching and removal of SiO_2 residues.

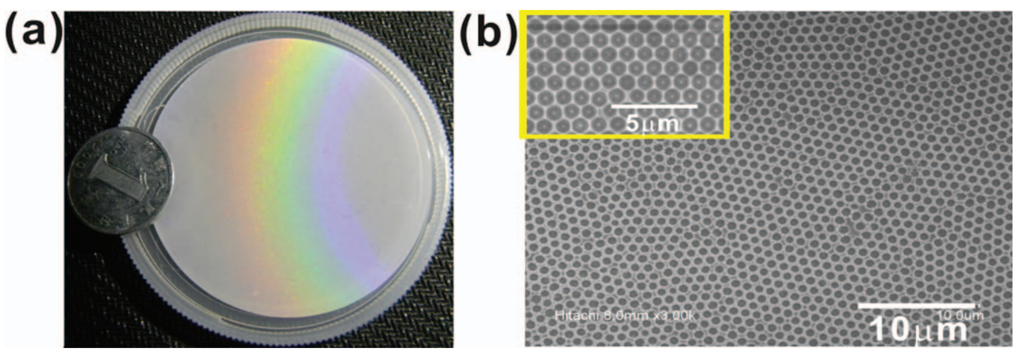

FIG. 2. (a) SiO2 nanobowls on a whole two-inch epitaxial wafer acted as an etching mask and (b) ITO nanobowls achieved after the ICP etching. The inset shows the SiO2 nanobowls mask on LED wafers before ICP etching process.

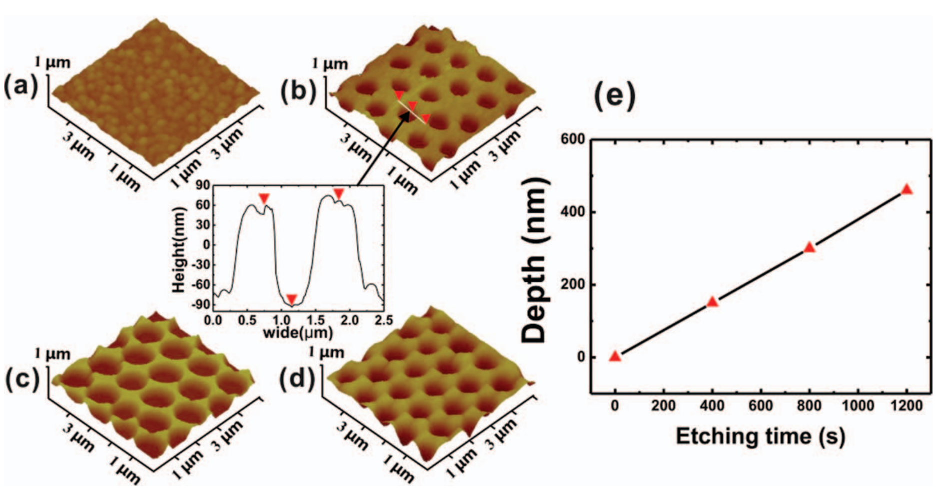

FIG. 3. 3D AFM images of (a) planar ITO, (b)–(d) nanobowls PhC with etching for 400s, 800s, and 1200s, respectively, and (e) the depth of nanobowls depending on ICP etch time. Inset image depicts the depth of nanobowls for 400s.

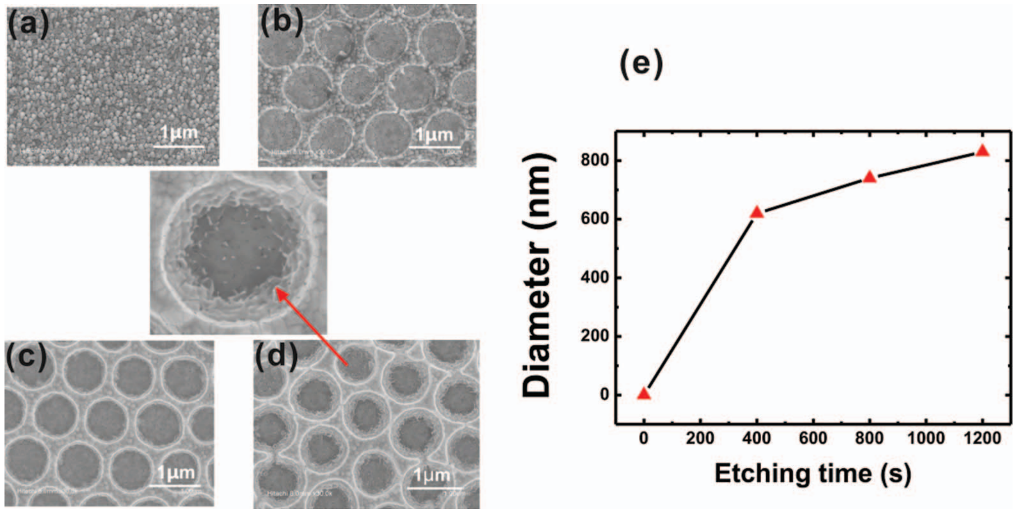

FIG. 4. SEM images of (a) planar ITO, (b)–(d) ITO nanobowl PhC structures etched for 400, 800, and 1200s, respectively, and (e) plot of the diameters of nanobowls depending on the ICP etch time.

to be 150 nm, 300 nm, and 460 nm for 400 s, 800 s, and 1200 s, respectively, as shown in Fig. 3(e). The ITO film beneath the deepest nanobowls (etched for 1200s) has been intentionally retained at 50 nm to perform as transparent conductive electrodes, providing low-resistivity current spreading ability in order not to damage the electrical performance of PhC-LEDs.[8] Scanning electron microscopy (SEM) images obtained from Hitach S4800, as illustrated in Fig. 4(a)–4(d), offer a wider view of the nanobowls on ITO layer. From Fig. 4(e), the diameters of nanobowls etched for 400s, 800s, and 1200s are evaluated to be approximately 620 nm, 740 nm, and 830 nm, respectively. It's clear to see that, for longer etching time beyond 1200s, the PhC structural integrity of ITO nanobowls will be destroyed.

Electroluminescence (EL) measurement of packaged and un-encapsulated PhC and conventional LEDs are carried out by collecting the emitted light with an integrating sphere optically coupled with a radiometrically calibrated spectrometer. The light output power-current (*LOP-I*) characteristics of LEDs with and without PhC structure are shown in Fig. 5, from which it is obvious to see that the PhC LEDs exhibit strong improvement in light emission over the unpatterned LEDs. At an injection current of 350 mA, the LOP of PhC-LEDs with nanobowls etched for 400s, 800s, and 1200s can be enhanced by 25.7%, 44.6%, and 63.5%, respectively, compared to that of conventional ones. This enhancement is due to the fact that ITO nanobowls will eliminate the total internal reflection effect at the ITO interface, and thus improve the light extraction from LEDs.[17] For ITO ($n_{ito} = 1.9$) and air ($n_{air} = 1$) interface, the critical angle of the total internal reflection is 31.8°. The light exceeding the critical angle will be reflected at the interface between air and ITO. Afterwards, the light is reflected back and forth in the dielectric film, and transformed into thermal energy through absorption. In order to extract this part of light, the periodic structures were formed and provided a vector $G = (2\pi/\Lambda)\,\hat{e}_{\Gamma K}$ in the tangential direction. Here, Λ is the period of the periodic texture. The in plane wave vector of K is changed to K′, which satisfies the equation of $K = K' + mG$, where m is an integer. Moreover, the wavelength of the emitting light must satisfy the condition of maximum diffraction efficiency of $\Lambda/\lambda \geq 2/3$.[18] Therefore, the extraction efficiency of light can be improved by changing the in plane wave vector, which couples the guided modes into radiation modes.[19] For the purpose of getting the angular emission profiles of the LEDs with/without PhC structure, the chips have been located on an aluminum supporting substrate, and bonded with Au-wires without epoxy encapsulation. As depicted in the far-field radiation patterns of Fig. 6(a), it is found that the full-width-at-half maximum (FWHM) is 165.7°, 154.9°, 148.6°, and 136.9°, for reference LEDs and PhC LEDs etched for 400s, 800s, and 1200s, respectively. The LEDs etched for 1200s is found to produce the most significant focus effect with respect to the reference samples, demonstrating

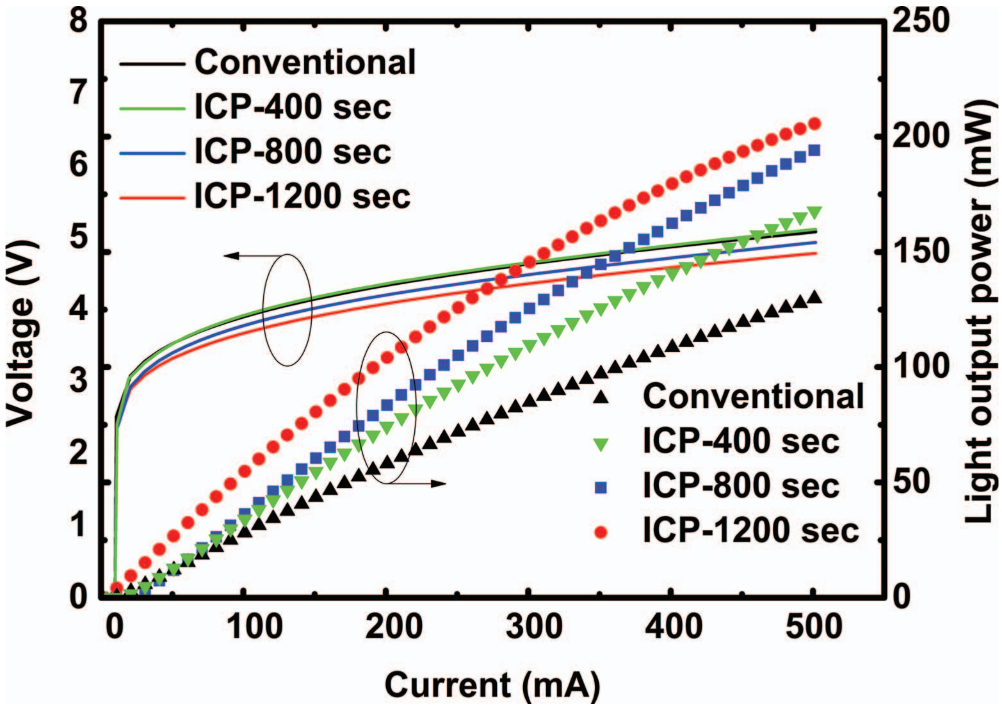

FIG. 5. *LOP-I-V* curves of reference LEDs and LEDs with nanobowls etched for 400, 800, and 1200s, respectively.

a reduction of 28.8° in the divergence angle. This is due to the nanobowls in the ITO layer help to confine the light to radiate from the vertical direction, which is unlike the random scattering in the result of the irregular surface roughening.[20] And this has proved that some waveguide modes in LEDs with ITO nanobowls PhC structures have been turned into radiation modes. On the other hand, from the measured *I-V* curves, the forward voltages for conventional LEDs, PhC LEDs etched for 400s, 800s, and 1200 sec are 3.25 V, 3.26 V, 3.13 V, 3.08 V, respectively, at a driving current of 20 mA. The slopes of the *I-V* curves in the linear region are also very similar, demonstrating that PhC structures on the top of ITO layer have little impact over the electrical performance of LEDs.

In order to further evaluate the effect of nanobowls on the ITO layer on the LEE enhancement in PhC-LEDs, a two-dimensional (2D) FDTD simulation has been implemented. Considering the in plane isotropy of hexagonal ITO nanobowls, the simulation model is simplified from 3D to 2D for saving the calculation time. The simulated LED structure is simplified, which consists of a 150 μm sapphire substrate, a 4 μm n-GaN layer, a 120 nm InGaN/GaN multi quantum wells (MQWs), and a 150 nm p-GaN layer, followed by a 510 nm ITO nanobowls with different size on the basis of the SEM and AFM images in Fig. 3 and Fig. 4, in comparison with an unpatterned one as well. The radiation source a point dipole polarized along the x, y, and z direction is placed at the middle of MQWs. The enhancement factor with different PhC structure has been plotted in Fig. 6(b), indicating the LEE of LEDs etched for 1200s is 57.3% higher than that of reference sample, which is a little smaller than that from experimental result. This is probably due to the fact that we are not taking into account the enhanced light scattering effect caused by a lot of facets on the surface of nanobowls, as shown in Fig. 4(d). Figure 6(a)–6(f) illustrate the propagation of electromagnetic waves passing through the ITO nanobowl PhC structures as well as the planar ITO surface. It is noted that, due to PhC structure suppressing lateral guiding modes and diffracting the optical energy to radiation modes, the propagation above the surface is highly improved for ITO nanobowl PhC structures than the conventional ones. Furthermore, LEDs etched for 1200s exhibits the most distinct converging effect and the field density under the PhC structure is much weaker compared to the case of other

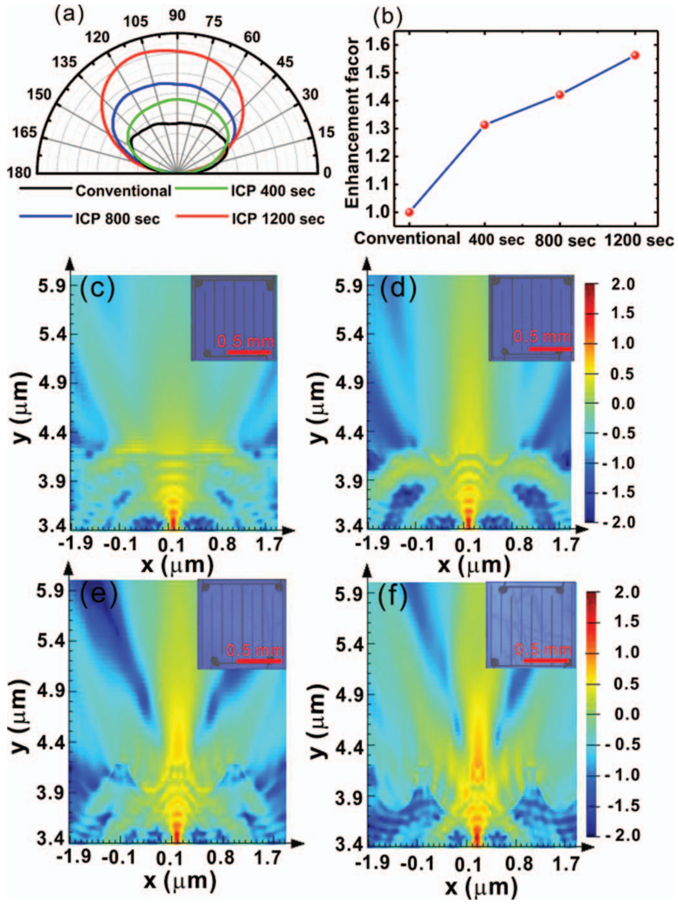

FIG. 6. (a) Far-field emission patterns of reference and PhC-LEDs, (b) FDTD simulated enhancement factor, (c)–(f) light propagation in the reference LED and LEDs etched for 400, 800, and 1200s, respectively. The inset shows the corresponding optical microphotographs of the above LEDs operated at an injection current of 2 mA.

LED samples with different etching times and without PhC structures. It is owing to coupling guided modes into normal direction, which is consistent with the result of far-field emission patterns in Fig. 6(a). As for the inset of Figure 6(c)–6(f), it shows the plane-view microphotographs of LEDs with different PhC structures operated at 2 mA etched for 400s, 800s, and 1200s, respectively, together with the reference sample with a planer ITO layer. Because more guided photons are redirected out of the active layer, the PhC-LED etched for 1200s appears brighter emission compared with the others.

IV. CONCLUSIONS

In conclusion, InGaN-based LEDs with different sizes of ITO nanobowl PhC structures on p-GaN layer have been proposed and demonstrated. Under an injection current of 350 mA, the improvement in light output power has reached about 63.5%, compared with the reference LED with a planar ITO layer. In addition, the measurements of far-field radiation patterns also show that the dispersion angle of nanobowl patterned ITO LEDs etched for 1200s (136.9°) are much narrower than that of planar ITO LED (165.7°). The finite-difference time-domain (FDTD) simulation has been also performed to further reveal the emission characteristics of PhC-LEDs, which is well consistent with experimental results.

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