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Enhanced light extraction efficiency of GaN-based light-emitting diodes with ZnO nanorod arrays grown using aqueous solution

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We report a dramatic increase in the light extraction efficiency of GaN-based blue light-emitting diodes (LEDs) by ZnO nanorod arrays on a planar indium tin oxide (ITO) transparent electrode. ZnO nanorods were grown into aqueous solution at the low temperature of 90 °C. With 20 mA current injection, the light output efficiency of the LED with ZnO nanorod arrays on ITO was increased by about 57% with no increase in a forward voltage over the conventional LEDs with planar ITO. The increased light extraction by the ZnO nanorod arrays is due to the formation of sidewalls and a rough surface, resulting in a multiple photon scattering at the LED surface. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3077606]

An increase in the light extraction efficiency of GaN-based light-emitting diodes (LEDs) is one of the most important issues in solid-state lighting. Several approaches, including photonic crystals,[1,2] conductive omnidirectional reflectors,[3,4] and surface roughening of indium tin oxide (ITO) transparent electrodes,[5–8] have been intensively studied. In particular, improving the LED efficiency by surface texturing and patterning on transparent electrodes, which can reduce light trapping caused by differences of refractive index has attracted much attention. For example, the output power of LEDs could be enhanced by 28% by texturing of the transparent *p*-electrode or by 45% through the use of textured ITO or Ga-doped ZnO thin layers as transparent *p*-electrodes.[5,9] However, these surface texturing methods involve expensive and energy consuming processes, long fabrication time using techniques such as lithography, laser holography, and imprint approaches, and also a dry etching process which could cause degradation of the electrical properties of the transparent *p*-GaN electrode.

Recently, vertically aligned ZnO nanorods/nanotips have been grown on the transparent electrode of GaN LEDs using metal-organic chemical vapor deposition (MOCVD) to improve the light output. Compared with conventional GaN LEDs, the output power of GaN LEDs with ZnO nanorod/nanotip arrays can be enhanced by up to 50% or 1.7 times.[10,11] Although the methods did not include either a dry etching process or complex lithography, the forward voltage (V_f) of GaN LEDs with ZnO nanorods/nanotips was increased significantly due to thermal damage of the transparent *p*-electrode during ZnO nanorod/nanotip growth at high temperature. These methods may therefore be unsuitable for a commercial device fabrication process, because this increase in V_f cannot improve the LED efficiency even though the output power of the LED is increased.

We report here a dramatic increase in the light extraction efficiency of GaN-based blue LEDs without degradation of the *p*-electrodes by dry etching and thermal damage. This method involves the self-assembly of ZnO nanorod arrays by a simple aqueous solution route, at the low temperature of 90 °C on the transparent ITO *p*-contact layer. This is a simple and effective process for improving the output power of GaN LEDs. Compared with conventional LEDs with only planar ITO, the light output efficiency of the LEDs with ZnO nanorod arrays on ITO was increased by up to 60% at 100 mA without degradation in the V_f.

The GaN-based LED epilayers were grown on sapphire substrates with *c*-face orientation by MOCVD under a reactor pressure of 200 Torr. The device structure of the InGaN multi-quantum-well (MQW) LEDs is *p*-GaN (0.25 μm, $p = 4\times10^{17}$ cm^{-3})/a five-period InGaN MQWs/*n*-GaN (1.5 μm, $n = 1\times10^{18}$ cm^{-3})/undoped GaN/sapphire. We fabricated InGaN/GaN MQW blue LEDs with conventional planar ITO (C-LED) and ZnO nanorod array/ITO (NR-LED) transparent *p*-contacts. We first partially etched the surfaces of the grown LED samples until the *n*-type GaN layer was exposed. Next, a Ti/Au (50/200 nm) layer as the *n*-type electrode was deposited by an e-beam evaporation technique. To achieve Ohmic contact of the *p*-GaN layer, the *p*-contact transparent copper indium oxide/ITO electrode (3/400 nm thick) was deposited onto *p*-GaN by e-beam evaporation and was annealed at 600 °C at O_2 ambient for 1 min. The bonding pad electrode of Cr/Au (50/200 nm) on the top surface of the ITO transparent electrode without ZnO nanorod was deposited by e-beam evaporation.

To realize ZnO nanorod arrays on the ITO electrode of the *p*-GaN layer, the ITO area of the *p*-electrode was made open by a photoresist lift-off process. A ZnO seed layer on

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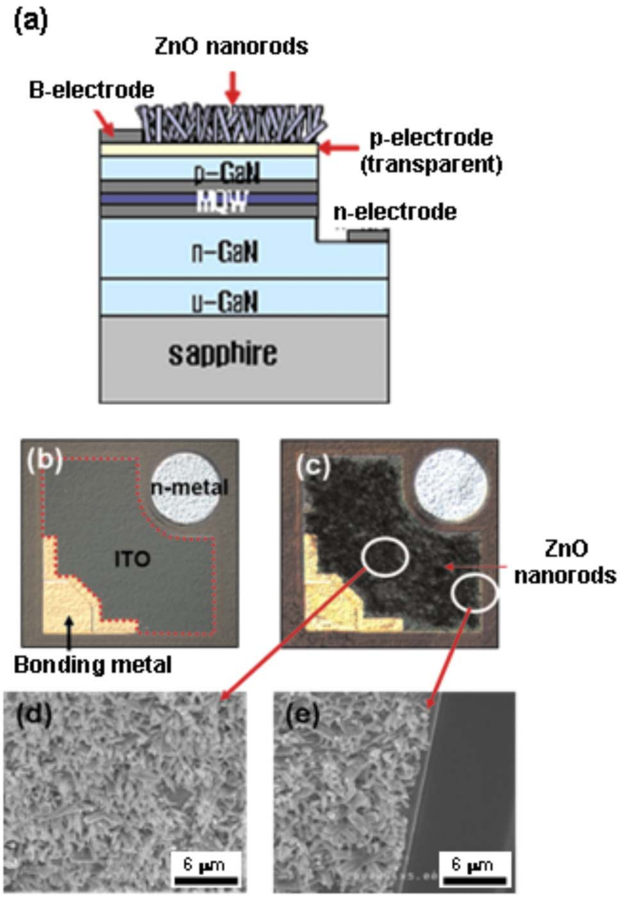

FIG. 1. (Color online) (a) Schematic diagram of GaN LED structure with ZnO nanorod arrays. (b) Top view micrograph image of C-LED and (c) top view micrograph image of NR-LED with FE-SEM images [d and e] of ZnO nanorods on the ITO of the *p*-GaN electrode.

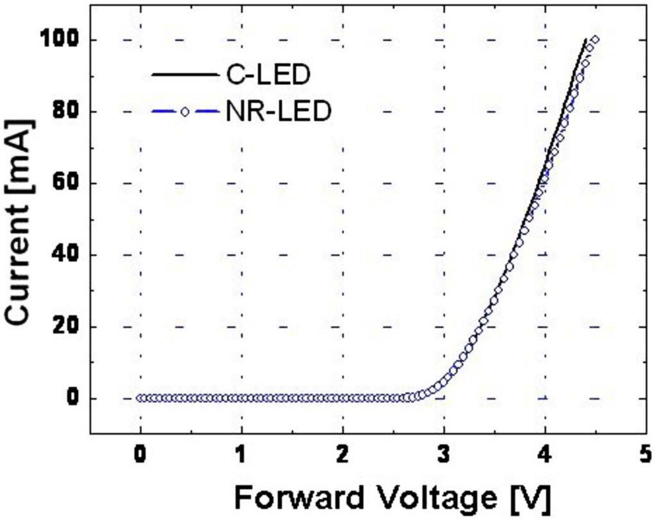

FIG. 2. (Color online) *I*-*V* curves of C-LED and NR-LED.

the *p*-GaN template was formed by a simple dipping process of the template for 5 min into a solution containing 5 mM zinc acetate $[Zn(C_2H_3O_2)_2]$ dissolved in de-ionized (DI) water at 90 °C. Following a seed-layer formation, the sample was preannealed for 5 min at 100 °C prior to main growth. ZnO nanorod arrays on nanosized ZnO seeds dispersed over the ITO electrode were grown by the "dipping-and-holding" process of the substrates into a solution consisting of DI water, 25 mM zinc nitrate hexahydrate $[Zn(NO_3)_2 \cdot 6H_2O]$, and 25 mM hexamethylenetetramine $[C_6H_{12}N_4]$ for 30 min at 90 °C.[12] To grow ZnO nanorods on the ITO of the *p*-electrode, the ITO electrode of the LED was bare by a lift-off process using photoresist. Well arrayed ZnO nanorods were selectively grown on the ITO surface of the *p*-electrode.

Electrical and optical characteristics of the LEDs were measured using an on-wafer testing configuration, comprising a parameter analyzer and optical detectors mounted above the LED chips. The optical detector is composed of optical fiber (1 mm in diameter) inserted in objective lens and the spectrum analyzer (Instrument Systems, CAS 140B). In addition, measurement angle of LED output power is tilted about 30° away from the surface normal.[13]

Figure 1 shows a schematic NR-LED structure with ZnO nanorod arrays grown on the top surface of the transparent ITO electrode [Fig. 1(a)], and plan-view images of the C-LED and the NR-LED [Figs. 1(b) and 1(c)]. As shown in Fig. 1(c), we grew the ZnO nanorod arrays on the ITO surface using photolithography patterning. The tilting-view field-emission scanning electron microscopy (FE-SEM) images in Figs. 1(d) and 1(e) show the general morphology of the ZnO nanorod arrays grown on the transparent ITO electrode, having uniform size distribution with an average length and diameter of 1 μm and 100 nm, respectively. Furthermore, the ZnO nanorods have excellent selectivity properties when grown on the ITO surface.

It is necessary that these ZnO nanorods grow without damage of the electrical properties of the LED by heat or plasma during nanorod growth. NR-LEDs and nanopatterned LEDs using ZnO nanorods and film have recently been reported.[10,11] To increase the LED efficiency, the output power must increase with constant V_f or smaller V_f. However, these results showed that the current-voltage (*I*-*V*) property was degraded due to *p*-electrode damage by growth of a ZnO film or ZnO nanorod at high temperature. The efficiency of the resulting LEDs was reduced as a result of increased V_f, although the output power of the LED was increased with better light extraction after exploiting ZnO nanorod/patterning to LEDs. In this regard, we propose a wet process to grow nanorods without damage of the *p*-electrode in LEDs. The wet process is a low temperature process so that the process gives no damage to the *p*-electrode and does not increase the V_f of the LED. It is therefore a very effective method that can generate nanostructures suitable for increasing light extraction.

Figure 2 shows *I*-*V* characteristics of the two fabricated LEDs. The V_f of the NR-LED was slightly larger than that of the C-LED. However, the LED operating voltage at the injection current of 20 mA shows no significant differences between the C- and NR-LEDs at 3.369 and 3.373 V, respectively. At 100 mA, the voltages of the C- and NR-LEDs are 4.41 and 4.50 V, respectively. The slightly larger V_f from the NR-LED is presumably because the undoped ZnO nanorods on the planar ITO are of high resistance. However, the introduction of the ZnO nanorod arrays would not result in significant degradation of the electrical properties of the LEDs given only a small increase in the V_f.

The enhanced light extraction efficiency of the NR-LEDs over the C-LED is mainly due to the formation of a textured surface on the NR-LED by ZnO nanorod growth. In

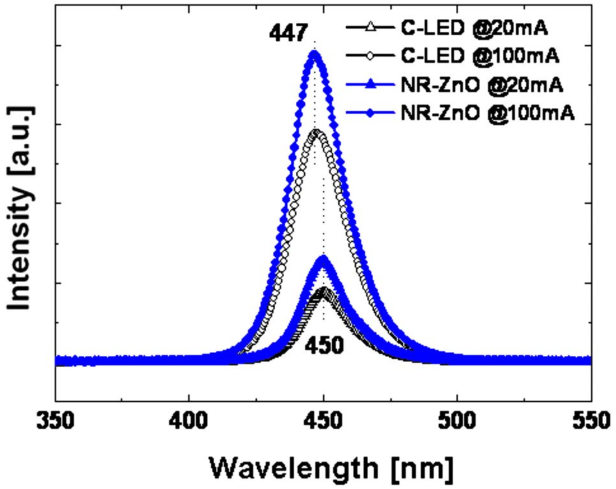

FIG. 3. (Color online) EL spectra of C-LED and NR-LED at 20 and 100 mA.

the NR-LED with a large number of nanorod sidewalls and a rough surface, photons generated in the MQW region should experience multiple scattering at the LED surface and can readily escape from the device. In other words, the ZnO nanorod arrays introduced via the simple aqueous solution route, leads to a dispersed angular distribution of the photons generated in the active layers, resulting in a larger escape cone for the photons in the NR-LED than in the C-LED. Furthermore, ZnO has a higher refractive index (about 2.0) than air or resin; also improving the light extraction efficiency.

Figure 3 shows electroluminescence (EL) spectra of the fabricated C- and NR-LEDs at injection currents of 20 and 100 mA. There are no significant differences in the EL peak positions (at 450 nm) of the two LEDs with the same full width at half maximum of 20 nm. However, EL intensities obtained from the NR-LED were larger than those achieved from the C-LED at injection currents of 20 and 100 mA. The ZnO nanorod density of NR-LED was controlled from low to high densities. In the case of low density nanorods, the light output of the NR-LED is about 15.9% and 21.0% higher than that of the C-LED at injection currents of 20 and 100 mA, respectively. At high density, relative to C-LEDs, the light output power of NR-LED is greater by 56.8% and 59.1% at 20 and 100 mA, respectively. This is shown in Fig. 4, which displays the current-light output power characteristics of the two fabricated LEDs. We suggest that these observations are attributed to the enhanced light extraction efficiency of the textured surface in the NR-LED with ZnO nanorod arrays.

In summary, we fabricated GaN-based blue LEDs with ZnO nanorod arrays on a planar ITO transparent electrode. We propose a wet process for nanorod growth without damage of the *p*-electrode in the LED, because this is a low temperature process; also there is no increase in V_f. At 20 mA current injection, the light output efficiency of NR-LED was greater by about 57% than a C-LED with only planar ITO. It is suggested that the enhanced light extraction by the ZnO nanorod arrays is due to the formation of a large number of nanorod sidewalls and a rough surface, resulting in a larger escape cone for the photons in the NR-LED than in the C-LED.

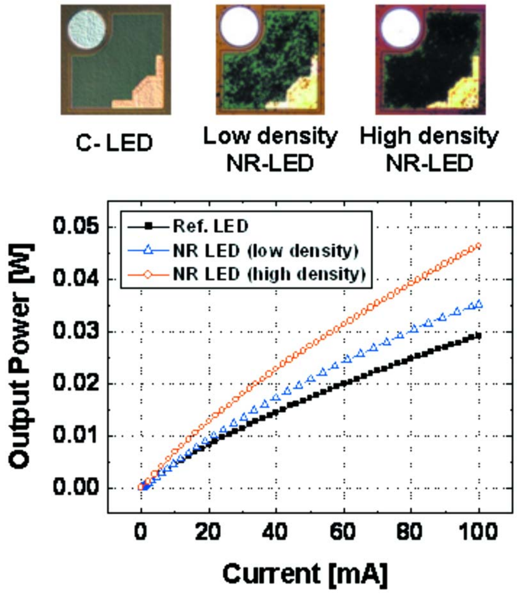

FIG. 4. (Color online) Micrograph images of C-LED, low density NR-LED, and high density NR-LED. Light output power of individual LEDs as a function of injection currents.

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