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Enhanced light output of GaN-based light-emitting diodes with ZnO nanorod arrays

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We report enhanced light output of GaN-based light-emitting diodes (LEDs) with vertically aligned ZnO nanorod arrays. The ZnO nanorod arrays were prepared on the top layer of GaN LEDs using catalyst-free metalorganic vapor phase epitaxy. Compared to conventional GaN LEDs, light output of GaN LEDs with the ZnO nanorod arrays increased up to 50% and 100% at applied currents of 20 and 50 mA, respectively. The source of the enhanced light output is also discussed. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2903153]

Considerable research has been recently devoted to the fabrication of high-brightness light-emitting diodes (LEDs) for general illumination.[1] The internal quantum efficiency of GaN LEDs, which affects brightness, typically exceeds 70%, much higher than the 10%–25% efficiency of conventional light sources such as light bulbs, incandescent electric lamps, and fluorescent lamps.[2] Nevertheless, the total light-output efficiency of LEDs is reduced to a few percent by the low light-extraction efficiency (LEE) due to the total internal reflection of the light generated in the LED active layers.[3] Accordingly, research has focused on methods of increasing the LEE of photons generated in the active layers, including LED dies,[4] flip-chip LEDs,[5] surface texturing,[6,7] and two-dimensional photonic crystals,[8] as well as on overall LED efficiency. However, the methods require complex and expensive processes or may result in surface damage.[9] Here, we report significantly improved light-output performance by growing vertically aligned semiconductor nanorod arrays on a top layer of GaN LEDs.

Since ZnO and GaN materials have similar refractive indices ($n=2.1-2.5$), the Fresnel reflection between GaN and ZnO is significantly reduced; thus, ZnO nanorod arrays have been employed in semiconductors to enhance the light-output efficiency of InGaN/GaN multiple quantum well (MQW) LEDs. In addition, as the band gap energy of ZnO is higher than the energy of the light emitted from InGaN/GaN MQW layers and the lattice constant misfit of 1.9% is small,[10] it is possible to heteroepitaxially grow high-quality ZnO nanorods on GaN, thereby ensuring a clean interface between ZnO and GaN and preventing light absorption in ZnO nanorods. When the light generated from MQWs is incident to the nanorods, it is easily extracted through side facets and the tip of the nanorods. Thus, the growth of ZnO nanorod arrays on GaN LEDs will increase the LEE of the LEDs.

Figure 1(a) is a schematic of a GaN LED with vertically aligned ZnO nanorod arrays. InGaN/GaN MQW LEDs, showing a dominant electroluminescent (EL) peak at a wavelength of 460 nm, were grown on the *c*-plane sapphire substrates by metalorganic vapor phase epitaxy (MOVPE) as reported previously.[11] Briefly, after the GaN LED substrates were cleaned with acetone and methanol in an ultrasonic bath, vertically well aligned ZnO nanorod arrays were heteroepitaxially grown on the top layer of the LEDs by catalyst-free MOVPE. Diethylzinc and oxygen, at flow rates of 0.5–9 SCCM (SCCM denotes cubic centimeter per minute at STP) and 20–100 SCCM, respectively, were employed as the reactant gases.[12] The surface morphology of the grown ZnO nanorods was investigated using field-emission gun-scanning electron microscopy. As shown in Fig. 1(b), the ZnO nanorods exhibited a uniform distribution in both diameter (~40 nm) and length (~1 μm), with a density as high as $10^{10}/cm^2$. The inset of Fig. 1(b) shows the morphology of a ZnO nanorod on top of a Ni/Au metal electrode. Note that the nanorod arrays on Ni/Au are vertically well aligned, with diameters, lengths, and density similar to those of ZnO nanorods grown on the open areas of the LEDs.

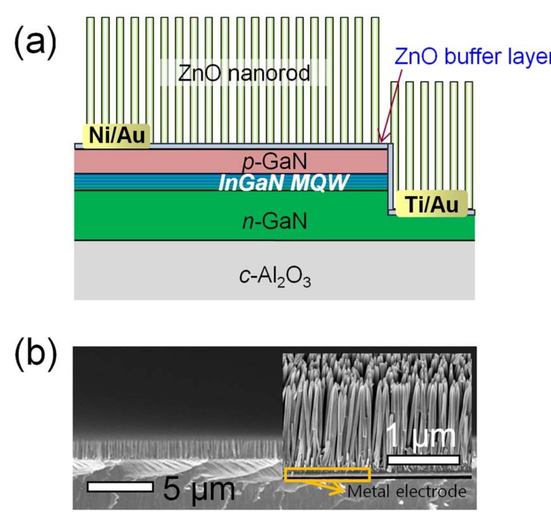

FIG. 1. (Color online) (a) Schematic of a GaN LED with vertical ZnO nanorod arrays. (b) FEG-SEM image of ZnO nanorod arrays grown on a GaN LED. ZnO nanorods are well aligned and are perpendicular to the GaN LED surface. The inset shows the morphology of a ZnO nanorod on top of a Ni/Au metal electrode, which are vertically well aligned, and the diameter/length/density is similar to that of ZnO nanorods in the open areas of the LEDs.

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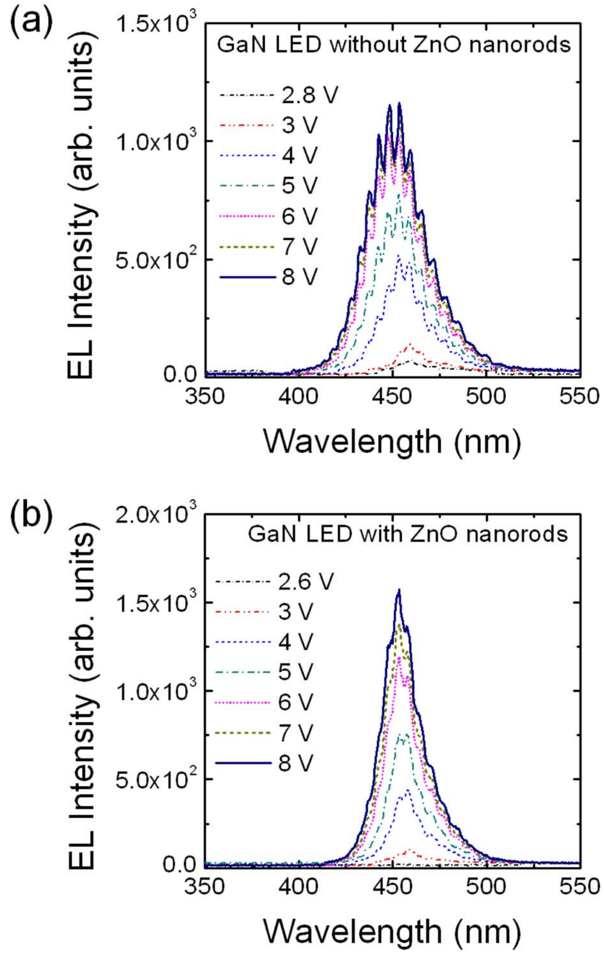

FIG. 2. (Color online) Room temperature EL spectra of GaN LEDs (a) without and (b) with ZnO nanorod arrays. The peak wavelength of GaN LEDs containing ZnO nanorod arrays shows a small, blueshift from 457 nm at 1 mA to 454 nm at 80 mA, while the GaN LEDs without ZnO nanorods exhibit a significant blueshift from 459 nm at 1 mA to 450 nm at 80 mA. In addition, the EL spectrum of GaN LED with ZnO nanorod arrays shows a smaller full width at half maximum value of the dominant peak than conventional GaN LEDs without exhibiting any interference effect.

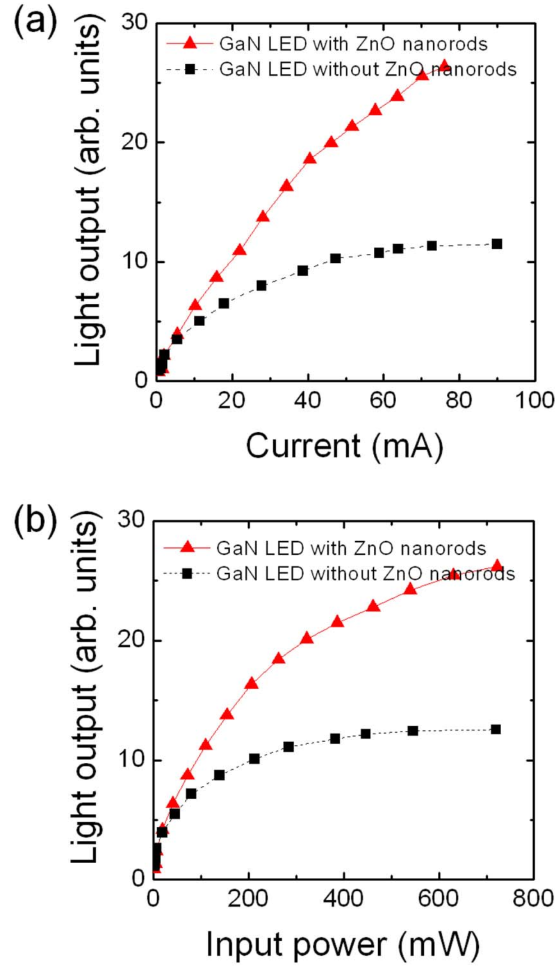

FIG. 3. (Color online) Light output power (L) as a function of (a) current (I) and (b) input power of GaN LEDs with and without ZnO nanorod arrays. The light output powers of the GaN LEDs with ZnO nanorods at currents of 20 and 50 mA were 50% and 100% higher, respectively, than those of the GaN LEDs without ZnO nanorods. Furthermore, the light output power of GaN LEDs without ZnO nanorods was almost saturated for currents above 60 mA, while GaN LEDs with ZnO nanorods showed no saturation up to 80 mA.

EL and current-voltage (I-V) curves for the LEDs were measured at various forward- and reverse-bias voltages at room temperature. The EL spectra were recorded using a monochromator and a photon-counting system equipped with a charge-coupled device, as detailed elsewhere.[13] Briefly, when a direct-current bias voltage was applied to the LEDs from a source meter, light was emitted from the front of the diodes through the ZnO nanorod array, and the EL intensities were recorded at various current levels at room temperature. ZnO nanorod arrays on the LEDs exhibited a transmittance of over 90% in the visible region with a sharp, fundamental absorption edge at 380 nm. To confirm the reliability of the fabrication process and the characteristics of the GaN LEDs, more than 30 LED devices were tested; they all exhibited similar optical and electrical characteristics.

The EL spectra of GaN LEDs, with and without ZnO nanorod arrays, as shown in Fig. 2, exhibited a dominant EL emission peak at 456 nm only. The absence of defect-related emission peaks strongly suggests that the growth of ZnO nanorods on GaN LEDs does not induce the formation of any radiative defects or damage on the GaN surface. With an increase in the applied current from 1 to 80 mA, the peak wavelength of GaN LEDs containing ZnO nanorod arrays exhibited only a small blueshift from 457 to 454 nm. In contrast, GaN LEDs without ZnO nanorods showed a significant blueshift from 459 to 450 nm, presumably as a result of band filling[14] and screening effects induced in the InGaN/GaN QW layers under a piezoelectric field.[15,16] The smaller blueshift in the EL peak position of GaN LEDs with ZnO nanorods is evidence that the InGaN/GaN QW layer has smaller internal field. Thus, we contend that the electrical field of GaN LEDs can be changed by growing ZnO nanorods on p-GaN, and that with the reduction in internal polarization, GaN LEDs with ZnO nanorods are very promising as high-efficiency LEDs. Furthermore, due to interference of the emitted light reflected by the sapphire/GaN and GaN/air interfaces and the smaller full width at half maximum value (18 nm) of the dominant EL peak compared to GaN LEDs without ZnO nanorods (25 nm), the EL spectrum of GaN LEDs with ZnO nanorod arrays exhibits greatly reduced multiple fringes. These results indicate that the ZnO nanorod arrays on GaN LEDs result in a decrease in the total internal reflection of the generated light in the active region of the LED.[17]

To further investigate the EL characteristics of GaN LEDs with ZnO nanorods, light output power (L) was mea-

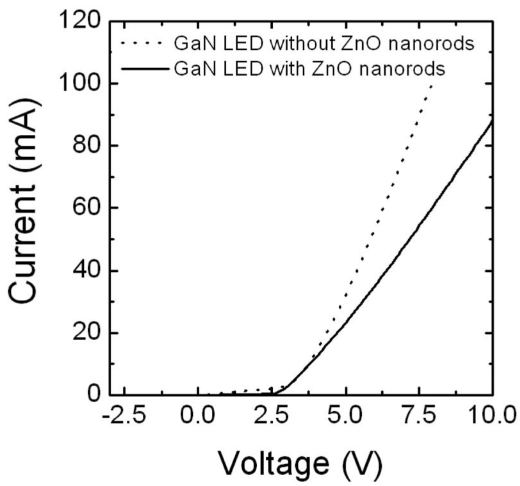

FIG. 4. *I-V* curves of LEDs with and without ZnO nanorods. Both *I-V* curves of the LEDs with and without ZnO nanorods exhibited nonlinear and good rectifying behavior with a turn-on voltage of 2.8 V and a leakage current of 1×10^{-5} A at −5 V. However, series resistance of the GaN LEDs increased from 50 to 83 Ω after ZnO nanorod growth.

sured as a function of current (I) and input power. Figure 3(a) shows *L-I* curves of GaN LEDs with and without ZnO nanorods. Since the experimental data were obtained from the same devices before and after ZnO nanorod growth on LED chips, other factors affecting the EL characteristics can be ignored. The *L-I* curves of both GaN LEDs, measured in front (top side through the ZnO nanorod arrays) of the devices, exhibited increased light output power with increasing applied current. As shown in Fig. 3(b), the light output power of the LEDs similarly increased with increased input power. In particular, the light output power of the GaN LED with ZnO nanorods exhibited a 50% increase at 20 mA, and furthermore, was not saturated up to 80 mA. The GaN LEDs without ZnO nanorods, however, exhibited an almost saturated light output power at currents above 60 mA. The greatly increased efficiency of the LEDs with ZnO nanorods at high current is a result of better heat dissipation through the nanorods due to both the higher thermal conductivity (1.47 W/cm K) of ZnO compared to sapphire (0.3 W/cm K) and the high surface/volume ratio of nanorod arrays. This is also confirmed by the small EL peak shift of the LEDs with ZnO nanorods due to the reduced device heating. Accordingly, the results strongly suggest that the GaN LEDs with ZnO nanorods exhibit a higher thermal stability as well as the increased LEE.

Electrical characteristics of the GaN LEDs were also determined from the *I-V* curves. Figure 4 shows *I-V* curves for the GaN LEDs with (solid line) and without (dotted line) ZnO nanorods on a *p*-GaN layer of the LEDs. Although the ZnO nanorods were subsequently coated on the *p*-GaN layer, the *I-V* curve of the LEDs with ZnO nanorods exhibited nonlinear and good rectifying behavior with a turn-on voltage of 2.8 V and a leakage current of 1×10^{-5} A at −5 V. From the *I-V* curves in Fig. 4, the series resistances of GaN LEDs with and without ZnO nanorods were estimated to be 83 and 50 Ω, respectively. The increase in series resistance of GaN LEDs with ZnO nanorods may originate from scattering at the ZnO/GaN layer interface. Also note that the operating voltage of the GaN LEDs with ZnO nanorods is higher than that of the LEDs without ZnO nanorods.

In conclusion, catalyst-free MOVPE of vertically aligned ZnO nanorods on GaN LEDs significantly increased their light output power. GaN LEDs with ZnO nanorod arrays had 50% enhanced light output power at an applied current of 20 mA compared to conventional GaN LED, which further increased to 100% at 50 mA, resulting presumably from increased LEE. This simple growth process for ZnO nanorods, without changing the LED structure, should be invaluable for increasing the fabrication yield and reducing the production costs of higher-brightness LEDs.

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