Enhancing light extraction of GaN-based blue light-emitting diodes by a tuned nanopillar array*

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(Received 20 June 2013; revised manuscript received 8 August 2013; published 3 December 2013)

Surface patterning of p-GaN to improve the light extraction efficiency of GaN-based blue light-emitting diodes (LEDs) has been investigated. Periodic nanopillar arrays on p-GaN have been fabricated by polystyrene (PS) nanosphere lithography; the diameter of the nanopillars can be tuned to optimize the electrical and optical properties of the LEDs. The electroluminescence intensity of the nanopillar-patterned LEDs is better than that of conventional LEDs; the greatest enhancement increased the intensity by a factor of 1.41 at a 20 mA injection current. The enhancements can be explained by a model of bilayer film on a GaN substrate. This method may serve as a practical approach to improve the efficiency of light extraction from LEDs.

Keywords: light-emitting devices, nanosphere lithography, electroluminescence, extraction efficiency

PACS: 85.60.Jb, 81.16.Nd, 78.60.Fi **DOI:** 10.1088/1674-1056/23/1/018502

1. Introduction

Solid-state lighting devices, which are now considered to be a new-generation light source, have developed rapidly in recent years due to its features of energy conservation, long service life, fast response time and small volume.[1] GaN based light-emitting diodes (LEDs) have been attracting a great deal of interest due to their capability to emit a spectrum from ultraviolet to green and their applications in traffic signals, displays and solid-state lighting. Consequently, there is widespread interest in improving the efficiency of LEDs. Generally, the light external quantum efficiency (EQE) of LEDs is decided by the light internal quantum efficiency (IQE) and the light extraction efficiency (LEE), $\eta_{\mathrm{EQE}} = \eta_{\mathrm{IQE}}\eta_{\mathrm{LEE}}$. Although the IQE of InGaN/GaN-based LEDs is reported to exceed 90%,[2] only a small fraction of light can be extracted because of the great difference in the refraction indexes (n) of GaN ($n = 2.5$) and air ($n = 1$).[3] According to Snell's law, the critical cone angle for light to escape from GaN to air is 23° and only about 4% of the light generated in the multiple quantum well (MQW) can escape into the surrounding space.[4] The majority of photons are reflected from the interface, become guided light in a waveguide formed by GaN and get reabsorbed. To increase the light extraction efficiency, various methods, such as roughed p-GaN surface,[5–9] roughed sapphire substrate,[10,11] photonic crystal[12–16] and patterned indium tin oxide (ITO) surface,[17–19] have been implemented in InGaN-based LEDs.

Most of the nanoscale patterning techniques, such as laser holography,[20] electron-beam lithography[21,22] and nanoimprinting lithography[14] are used to obtain nanoscale surface textures. Compared to the other techniques, nanosphere lithography[17] has the advantages of low cost and high throughput, and is very suitable for patterning LEDs. Currently, various monodisperse nanospheres (size variation $<$ 5%) can be obtained at low cost. Therefore, different periodic nanostructures can be formed to optimize the LEE of LEDs by nanosphere lithography.[23] Of course, in the same period, the diameter of the nanopillars can be tuned to optimize the LEE of the LEDs.[24–26] Compared to other nanospheres, the PS nanosphere is easily etched, so the diameter of PS nanospheres could be tuned to obtain nanopillars of different diameters. However, such research has rarely been reported.

In this work, periodic nanopillar arrays on p-GaN layer have been fabricated by polystyrene (PS) nanosphere lithography, and the diameter of the nanopillars is tuned by oxygen plasma etching and can be used to optimize the electrical and optical properties of LEDs. The electroluminescence intensity of the patterned LEDs is enhanced by a factor of 1.41 at 20 mA injection current compared to that of conventional LEDs. The details of the experimental procedure and the optical and electrical characteristics of the fabricated LEDs will also be discussed.

*Project supported by the National Natural Science Foundation of China (Grant Nos. 10774195, U0834001, 10974263, 11174374, and 10725420), the Key Program of Ministry of Education, China (Grant No. 309024), the Program for New Century Excellent Talents in University of Ministry of Education of China, and the National Basic Research Program of China (Grant No. 2010CB923200).

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2. Experimental methods

Metal-organic chemical vapor deposition (MOCVD) was used to grow GaN LED epilayers on 2-inch sapphire wafers for the fabrication of GaN LED chips. Figure 1(a) shows the entire LED epilayers structure, which consists of a 3.5-μm undoped GaN buffer layer (u-GaN), a 2.3-μm Si-doped n-GaN layer, a 3-period InGaN/GaN multiple quantum well (MQW) active layer and a 240 nm Mg-doped p-GaN layer. The wavelength of the emitting light is about 465 nm.

A patterned p-GaN contact layer with a nanopillar array was fabricated through nanosphere lithography; the process flow is shown in Fig. 1. First, a hexagonal close-packed monolayer of PS nanospheres with a 450-nm diameter was formed on the p-GaN layer. Second, oxygen plasma etching was used to reduce the diameter of nanospheres. By increasing the etching time, the diameter of the nanospheres was reduced. In order to adjust the diameter of the PS nanospheres, the orderly PS nanospheres were etched for 0 s, 7 s, 14 s, 19 s and 24 s, respectively. Then, the p-GaN layer with a monolayer mask of tailored PS spheres was etched by an inductively coupled plasma (ICP) etching machine via the gas flow of nitrogen with Cl_2/BCl_3. The etching rate is about 6.5 nm/s and the etching time is 22 s. Therefore, the corresponding etching depth is about 140 nm, which is of course the height of the nanopillars. Finally, the PS spheres were removed by trichloromethane solvent with sonication.

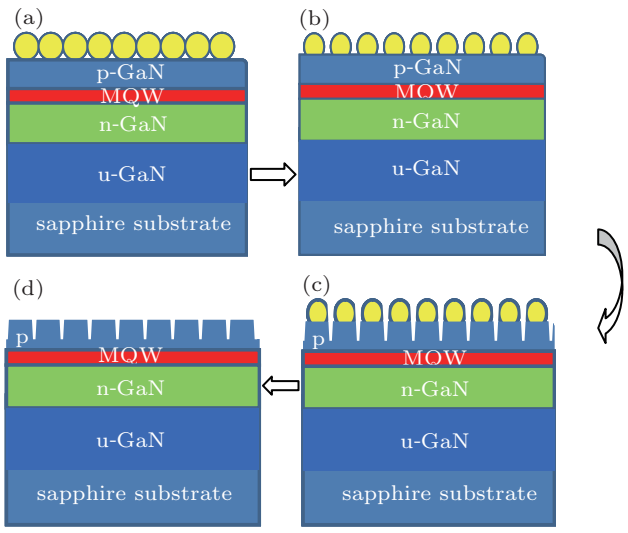


Fig. 1. (color online) Schematic of the fabrication process for a nanopillar array on a p-GaN surface in an LED. (a) Deposition of PS spheres on the p-GaN surface. (b) Etching using oxygen plasma to control the diameter of the PS spheres. (c) Etching by ICP to fabricate the nanopillar structure. (d) Removal of the PS nanospheres, retaining the nanopillar.

A transparent ITO electrode with a thickness of 400 nm was deposited onto the nanopillar-patterned p-GaN surfaces and conventional LEDs. The LED chips with dimensions of 300 μm×300 μm were made by mesa-etching the exposed n-type GaN via standard lithography, ITO wet etching and subsequent ICP etching. Cr/Pd/Au was deposited on the top ITO surface and the exposed n-GaN layer as a contact metal for both the p- and the n-GaN layers, as shown in Fig. 2. The surface morphologies of LED chips were examined by scanning electron microscopy (SEM). The optical output power–current (O–I) and current–voltage (I–V) measurements were carried out using a probe station system; the LED chips were not encapsulated during the electrical tests.

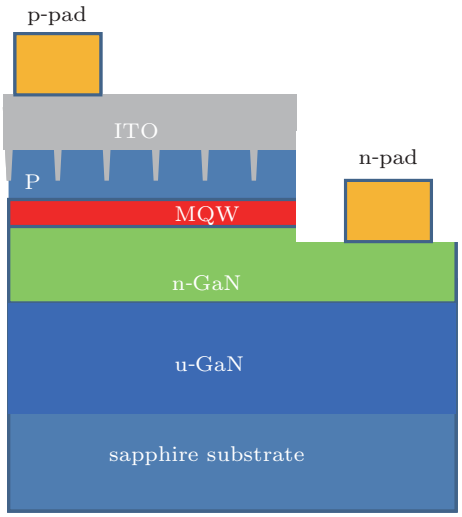


Fig. 2. (color online) Schematic illustration of a modified LED structure with a nanopillar-patterned p-GaN layer.

3. Results and discussion

3.1. Fabrication of a nanopillar array on the p-GaN surface

Six samples were fabricated in this work, including the conventional LEDs. Five samples with a nanopillar patterned p-GaN layer were marked as samples A, B, C, D and E, which were etched with oxygen plasma for 0 s, 7 s, 14 s, 19 s and 21 s, respectively. After a p-GaN layer was etched by ICP for 22 s, the PS nanospheres were removed and the pattern of nanopillars was formed on top of the p-GaN layer. Figure 3 shows SEM images of samples A, B, C, D and E.

As shown in Figs. 3(a)–3(e), when the PS nanosphere mask is etched with oxygen plasma longer, the diameters of the nanopillars become smaller. By measuring the SEM images, the diameters of nanopillars in samples A, B, C and D are found to be about 420 nm, 400 nm, 350 nm and 300 nm, respectively. The nanostructure of sample E is irregular since the PS masks were etched too much, so sample E was not considered further. The SEM images also show that the nanopillars are truncated nano-cones. The diameters of the top and bottom are different and the effective refractive index of the nanostructure shows a graded index profile. Figure 3(f) shows across sectional image of sample C and the height of the nanopillars are about 140 nm. The heights of the nanopillars of samples A, B, C and D are the same.

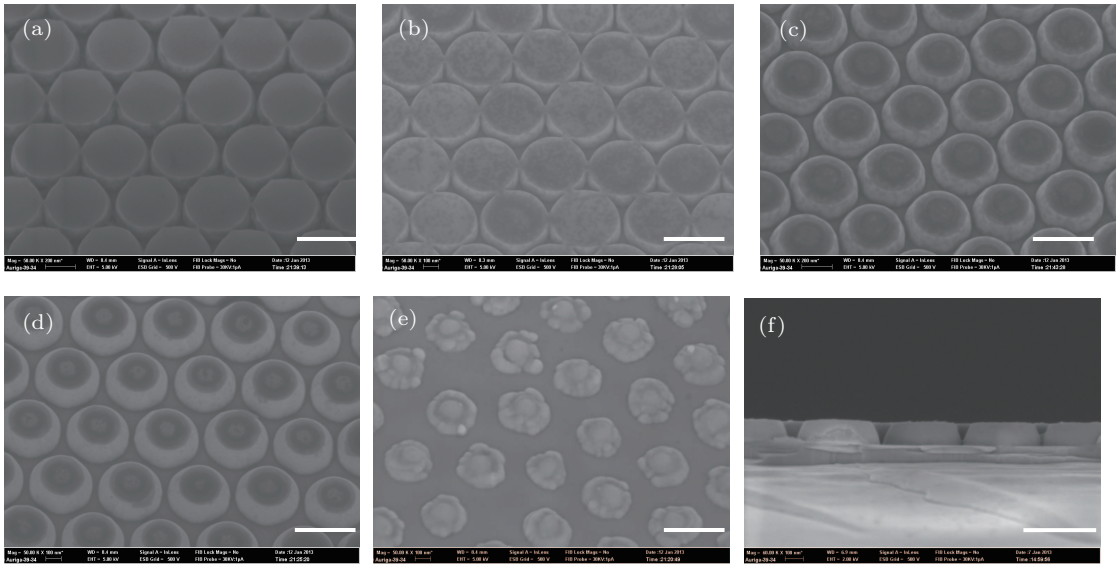


Fig. 3. SEM images of the nanopillar-patterned p-GaN layer. Scale bars are 400 nm. (a)–(e) 30° tilt images of samples (a) A, (b) B, (c) C, (d) D and (e) E, respectively. (f) Cross-sectional image of sample C. The PS nanospheres on the p-GaN layers of samples A, B, C, D and E were etched with oxygen plasma for 0 s, 7 s, 14 s, 19 s and 24 s, respectively. After ICP etching for 22 s, the PS spheres were removed and the fabrication of the nanopillar pattern on top of the p-GaN layer was complete.

3.2. Electroluminescence of the LEDs

By using above-mentioned standard fabrication processes, the samples A, B, C, D and a conventional LED were prepared. The electroluminescence intensity of sample D was very faint; this might be due to the fact that as the etched area became larger, the leakage current increased rapidly, so we do not consider sample D further. All of the devices were fabricated from the same LED wafer in order to eliminate the variations from wafer to wafer and ensure the reliability of the results. Figure 4(a) shows the optical output power versus injection current (*O*–*I*) for three nanopillar-patterned LEDs and a conventional LED. At the same injection current, the optical output powers of the three pillar-patterned LEDs were higher than that of the conventional LED. For instance, the optical outputs (at the injection current 20 mA) of the nanopillar-patterned samples A, B and C are about 13%, 33% and 41% greater, respectively, than that of the conventional LED. At the 100 mA injection current, the electroluminescence intensities of samples A, B and C are about 12%, 27% and 33% greater, respectively.

Figure 4(b) shows the forward current–voltage (*I*–*V*) characteristics of three pillar-patterned LEDs and a conventional LED. It is clear that the LED with a nanopillar-patterned p-GaN layer exhibits nearly the same *I*–*V* characteristics as the conventional LED. At the 20-mA injection current, it was found that the forward voltages of the patterned LEDs were in the range of 3.3–3.4 V, similar to that of the conventional LED.

The results show that the light extraction efficiency of the samples with nanopillar structures was enhanced and the greatest enhancement was 41% at an injection current of 20 mA. The causation can be explained from various points of view. First, the roughened surfaces can provide emitted photons with multiple chances to escape from the GaN surface. In detail, the roughened surface can redirect emitted photons, which makes the photons that were originally emitted out of an escaping cone go back into the escaping cone; therefore, the light extraction

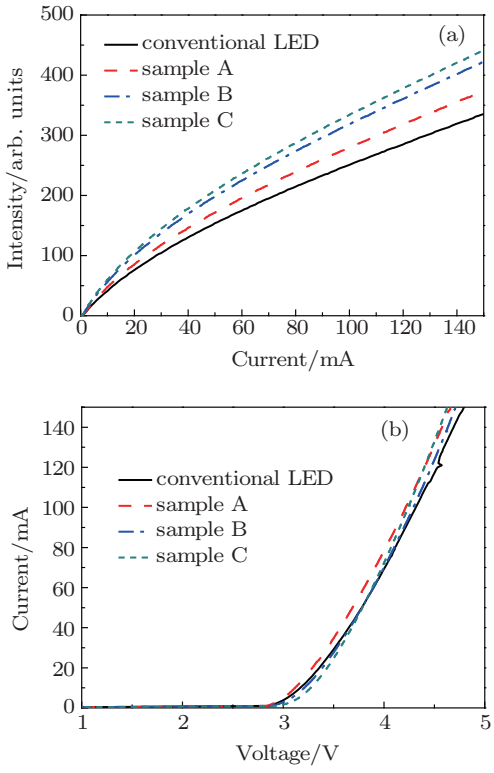


Fig. 4. (color online) Electroluminescence curves of three nanopillar-patterned LEDs and a conventional LED. (a) The optical output power versus injection current (*O*–*I*) characteristics. (b) The current versus voltage (*I*–*V*).

efficiency is improved.[17,27] Second, the dimension of the nanostructure is on the order of the light wavelength, so the periodic nanostructure serves as a two-dimensional grating. Such Bragg scattering assists the photons with multiple opportunities to escape from the p-GaN surface into the ITO interface.[5]

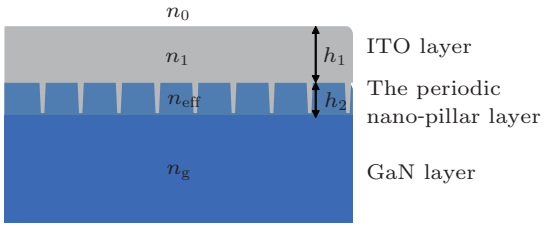


Fig. 5. (color online) Cross-sectional simplified illustration of the bilayer structure of an LED with nanopillars fabricated on a p-GaN layer.

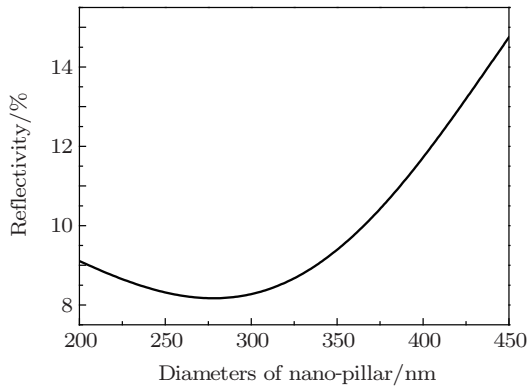


Fig. 6. Dependence of the reflectivity of a patterned p-GaN layer on the diameter of the nanopillars under effective medium theory.

Now, we turn to the effect of the nanopillars' diameter on the light extraction efficiency. The results show that the light output power of sample C is relatively high. This can be explained by a simple model, which is composed of bilayer film on GaN substrate as shown in Fig. 5. The first layer is ITO, which has a thickness of 400 nm. The second layer is a mixture of ITO and p-GaN due to the presence of the nanopillars and the thickness is about 140 nm. According to effective refractive index theory, the refractive index of this layer can be expressed as[28]

$$n_{\rm eff} = [n_{\rm GaN}^2 f + n_{\rm ITO}^2 (1-f)]^{1/2},$$

where $n_{\rm GaN}$ is the refractive index of GaN, $n_{\rm ITO}$ is the refractive index of ITO, f is the filling factor of p-GaN in a nanostructured layer. When the diameter of the nanopillars is tuned, the filling factor f also changes, which results in the change of $n_{\rm eff}$. At normal incidence, the reflectivity of the bilayer films could be calculated by the transfer matrix method, which is shown in Fig. 6. It can be found that the reflectivity of the bilayer film decreases with a decrease of the nanopillar diameter when the diameter is in the range of 300 nm to 450 nm. In experiment, the light extraction efficiency of patterned LEDs is increased with the decrease of the nanopillar diameter, which can be seen from the changes of the light output power of samples A, B and C. This calculated result is very consistent with the experimental result. In this case, the diameter of the nanopillar can be tuned to optimize the LEE of LEDs.

4. Conclusions

In summary, we have fabricated LEDs with a nanopillar-patterned p-GaN layer via nanosphere lithography. The diameter of the nanopillars can be tuned by oxygen plasma etching. The experimental results show that light extraction of nanopillar-patterned p-GaN LEDs is enhanced compared with conventional LEDs. The greatest enhancement was 41% at an injection current of 20 mA. The light enhancement can be attributed to angular randomization of photons by surface scattering and diffraction by the periodic nano-grating. The differing enhancement of light output for different diameters of nanopillars can be explained by a model of bilayer film on GaN substrate. The utilization of such low cost and controllable nanosphere lithography could serve as a practical approach to increasing the efficiency of light extraction from LEDs.

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