Strong light-extraction enhancement in GalnN light-emitting diodes patterned with TiO₂ micro-pillars with tapered sidewalls

Ming Ma, ¹ Jaehee Cho, ^{2,a)} E. Fred Schubert, ² Yongjo Park, ² Gi Bum Kim, ³ and Cheolsoo Sone ³

¹Future Chips Constellation, Department of Materials Science and Engineering, Rensselaer Polytechnic Institute, Troy, New York 12180, USA

²Future Chips Constellation, Department of Electrical, Computer, and Systems Engineering, Rensselaer Polytechnic Institute, Troy, New York 12180, USA

(Received 15 May 2012; accepted 17 September 2012; published online 2 October 2012)

An effective method to enhance the light extraction for GaInN light-emitting diodes (LEDs) is reported. The method employs TiO₂ micro-pillars with tapered sidewalls, which are refractive-index-matched to the underlying GaN. The tapered micro-pillars are fabricated by using reflowed photoresist as mask during CHF₃-based dry etch, with O₂ added in order to precisely control the taper angle. LEDs patterned with TiO₂ micro-pillars with tapered sidewalls show a 100% enhancement in light-output power over planar reference LEDs. The measured results are in good agreement with ray-tracing simulations, showing strong potential of optical surfaces that are controlled in terms of refractive index and lateral structure. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4756797]

One of the major obstacles facing light-emitting diodes (LEDs) is the occurrence of trapped light inside a highrefractive-index semiconductor. This phenomenon leads to a significant portion of the light remaining confined inside the semiconductor material. For example, only 4% of the light can escape from one surface of a planar GaInN LED.² Thus, the light-extraction efficiency of LEDs is severely limited in planar LEDs. A variety of methods have been reported to enhance the light extraction for LEDs, including LEDs with textured surfaces,^{3,4} LEDs with photonic crystals, 5,6 LEDs grown on patterned substrates, 7,8 and LEDs coated with patterned graded-refractive-index coatings. 9,10 A careful review shows that most of the above-mentioned approaches involve formation of characteristic features such as pillars, 9,10 holes 5,6 and hexagonal pyramids. 3,4 Although crystallographic wet chemical etching of a nitrogen-face (N-face) GaN surface can create hexagonal pyramids with a certain pyramid taper (e.g., $\theta \approx 58^{\circ}$ between the inclined plane and the base plane of the pyramid), 11,12 the pyramid taper cannot be arbitrarily selected. Furthermore, the engineering and fabrication of the sub-micron structures of a photonic crystal is elaborate and expensive. Thus, developing structures, controlled in refractive index and in lateral structure, is desirable. Indeed, the advanced control of both refractive index 13 and lateral structure 14 of an optical surface is highly promising and should be explored in greater detail.

One of the key characteristics of the LED surface features that affect the light extraction of LEDs is the feature's sidewall angle of inclination. Figure 1 illustrates the light extraction from a rectangular-parallelepiped-shaped LED with no surface features (Fig. 1(a)), from a rectangular-parallelepiped-shaped LED with a micro-pillar with vertical sidewalls (Fig. 1(b)), and from a rectangular-parallelepiped-shaped LED with a micro-pillar with tapered sidewalls

(Fig. 1(c)). For simplicity, the micro-pillars are refractive-

101, 141105-1

³Advanced Development Group, LED Business, Samsung Electronics, Yongin 446-920, Korea

index-matched to the LED semiconductor. As shown in Fig. 1(a), for a LED with no surface features, only those light rays striking the interface at an angle less than the critical angle, $\theta_{\rm c} = \sin^{-1} (n_{\rm air}/n_{\rm semiconductor})$, can be transmitted to the surrounding air. Thus, the transmittance of light through a planar surface is restricted to an escape cone, whose half angle is defined by the critical angle. When a GaN LED $(n_{\text{semiconductor}} \approx 2.5)$ is surrounded by air $(n_{\text{air}} = 1)$, the critical angle at the interface is only 24°. However, in case of a cylindrical micro-pillar with vertical sidewalls being formed on a semiconductor surface, as shown in Fig. 1(b), light is extracted through the top surface of the pillar as well as the sidewalls of the pillar, thus, enhancing the total range of angles which are extracted from the semiconductor. The light escape cone has now increased, because it encompasses both the original escape cone and the escape cones associated with the pillar sidewalls. If a LED is coated with a micro-pillar with tapered sidewalls, as shown in Fig. 1(c), the light escape cone is further increased. This is because, although the light escape cone associated with the top surface remains the same, the light escape cones associated with the sidewalls are now increased, i.e., more oblique light is directly out-coupled through tapered sidewalls than through vertical sidewalls. However, if the taper of the sidewalls keeps increasing, the escape cones associated with the top surface and the sidewalls will start to overlap. As a result, the total light extraction will saturate, and then decrease. Thus, the taper of the micro-pillars (the pillar taper) needs to be optimized. On the other hand, for the trapped light rays, a micro-pillar with tapered sidewalls located on a rectangular-parallelepiped-shaped semiconductor can redirect the trapped light rays more efficiently so that those light rays re-strike the interface at different angles until eventually being extracted; this results in more efficient light extraction. By contrast, a micro-pillar with vertical sidewalls

^{a)}choj6@rpi.edu.

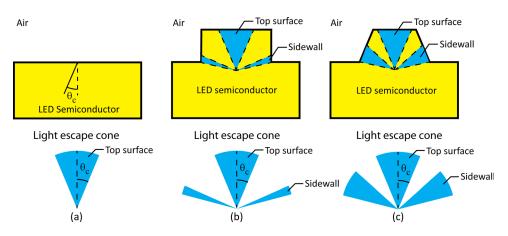


FIG. 1. Schematic diagrams of light extraction from (a) a rectangular-parallelepiped-shaped LED with no surface features, (b) a rectangular-parallelepiped-shaped LED with a micro-pillar with vertical sidewalls, and (c) a rectangular-parallelepiped-shaped LED with a micro-pillar with tapered sidewalls.

on a rectangular-parallelepiped-shaped semiconductor will have the trapped light rays re-strike the interface at either the same angle or its complementary angle. In other words, a micro-pillar with tapered sidewalls serves as a more efficient "redirection center" for the trapped light rays than a micropillar with vertical sidewalls. However, as the pillar taper keeps increasing, the ability of the micro-pillars to redirect the trapped light rays will also reach a maximum and then decrease, since a micro-pillar with too large a pillar taper will have its sidewalls almost parallel to the top surface of the semiconductor, and thus loses its functionality to efficiently redirect the trapped light rays. Therefore, to maximize light extraction, micro-pillars must be controlled in terms of their lateral sidewall structure. Thus, the taper of the micro-pillars (the pillar taper) plays a very important role in the light extraction of LEDs. Here the pillar taper is defined as

$$Pillar \ taper = \cot(\theta) = l/h, \tag{1}$$

where θ is the angle between the pillar sidewall and the LED top surface, as illustrated in the inset of Fig. 2, and l and h are also defined in the inset. According to this definition, a pillar with vertical sidewalls will have a θ of 90° and a pillar taper of 0; and the hexagonal pyramids created by crystallographic wet chemical etching of an N-face GaN surface will

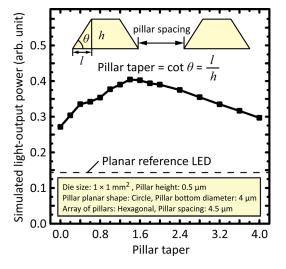


FIG. 2. Light-output power of a GaInN LED patterned with an array of refractive-index-matched micro-pillars, as a function of the pillar taper, simulated by ray tracing. The inset illustrates the definition of the pillar taper.

have a θ of 58° and a pillar taper of 0.6. Note that the pillar taper increases as θ decreases.

Figure 2 shows the simulated light-output power (LOP) as a function of the pillar taper for a GaInN LED patterned with an array of refractive-index-matched micro-pillars, as simulated by the ray-tracing method. In the simulation, the LOP is determined by counting extracted light rays through a spherical far-field receiver with infinite radius that is surrounding the whole LED die. The refractive index of the pillar is selected to be matched with that of GaN ($n \approx 2.5$). The absorption coefficient of GaN is assumed to be 10 cm⁻¹. A split-ray (reflection and transmission rays) method is employed and Fresnel loss at each interface is included. A total of 100 000 rays is used in each simulation. The LED die size is set to be $1 \times 1 \text{ mm}^2$. The micro-pillars used in the simulation have a circular planar shape, with a fixed height of $0.5 \,\mu m$ and a fixed bottom diameter of $4 \,\mu m$. The micropillars are arranged in a hexagonal array and the pillar spacing is kept at 4.5 μ m, as described in the inset of Fig. 2. A planar LED without surface features is simulated as a reference LED. Inspection of Figure 2 shows that for LEDs patterned with this micro-pillar-array structure, the LOP first increases with the pillar taper, and then decreases as the pillar taper further increases. The maximum LOP is achieved for micro-pillars with a pillar taper of 1.5, which corresponds to an angle θ of 34°. The ray-tracing-simulation results of Figure 2 also show that when compared with planar reference LEDs, LEDs patterned with micro-pillars with vertical sidewalls (taper = 0) can increase the light extraction. Moreover, LEDs patterned with micro-pillars with tapered sidewalls can further increase the light extraction of LEDs, and there is an optimum pillar taper for light extraction with regard to a specific micro-pillar structure. Therefore, a method that cannot only fabricate micro-pillars with tapered sidewalls, but also control the pillar taper, is highly desirable.

In this paper, we present an effective and elegant method to fabricate TiO_2 micro-pillars with tapered sidewalls and hence greatly enhance the light extraction for GaInN LEDs. TiO_2 micro-pillars with tapered sidewalls are fabricated by using reflowed photoresist as a mask during dry etch and by introducing O_2 , as additional etch gas together with CHF₃, with O_2 being added in order to precisely control the taper angle. Thin-film 1×1 mm² GaInN LEDs (N-face up) emitting at 445 nm and having no surface features are used as a reference device (planar reference LED). The vertical LED devices we used in our experiments have a chip design that is

very similar to the thin-film flip-chip geometry, ¹⁵ so the top N-face GaN surface is not covered by any metal electrodes. In addition, there is a layer of undoped GaN on top of the n-type GaN, which will protect the active region from dry etch damage. At first, about 500-nm-thick TiO2 is sputterdeposited on the N-face GaN surface of GaInN LEDs. TiO₂ is chosen since it is refractive-index-matched to GaN at the wavelength of 445 nm, so that there is minimal out-coupling loss (Fresnel reflection loss). The photo-lithography for patterning the TiO₂ films is performed using Shipley Company's S1813. Subsequently, the developed photoresist is hard-baked either at 120 °C for 2 min (the non-reflowed photoresist) or at 145 °C for 5 min (the reflowed photoresist). The TiO₂ film is then patterned using the photoresist as a mask during inductively coupled plasma (ICP) reactive ion etch (RIE). TiO₂ micro-pillars with vertical sidewalls are fabricated using the non-reflowed photoresist as a mask and using CHF₃ only during dry etch, with 600 W ICP power, 250 W RIE power, 60 sccm CHF₃ at 25 mTorr; the resulting micro-pillar is shown in Fig. 3(a). While TiO₂ micro-pillars with tapered sidewalls are fabricated using the reflowed photoresist as a mask and using the same etching conditions except introducing O2 as additional etch gas together with CHF₃ during dry etch. The resulting micro-pillars are shown in Figs. 3(b)-3(d). O₂ is introduced to etch the photoresist isotropically, while CHF₃ etches the TiO₂ anisotropically. As a result, TiO₂ micropillars can have tapered sidewalls. Furthermore, the pillar taper can be controlled by the flow rate of O_2 , which controls the etch rate of the photoresist. As shown in Fig. 3, the pillar taper first increases as the flow rate of O₂ increases, and then decreases as the flow rate of O2 further increases. As shown in Fig. 3(c), TiO₂ micro-pillars with a pillar taper of about 1.4 are achieved. And this pillar taper is close to the optimum pillar taper for this micro-pillar structure as predicted by the ray-tracing simulation. The LOP from the LED chip is measured using a lab-made far-field emission intensity measurement setup with a Si photo-detector that is moved by a rotating arm similar to a goniometer. The measured LOP is calculated after integration over all angles, and thus can be compared with the simulated LOP.

Planar reference LEDs, LEDs patterned with TiO_2 micro-pillars with vertical sidewalls (taper = 0), and LEDs

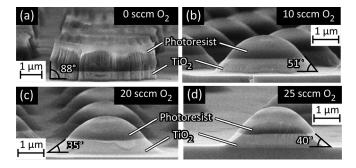


FIG. 3. Scanning electron micrographs of TiO₂ micro-pillars fabricated with different etch conditions: (a) with non-reflowed photoresist and 0 sccm of O₂, resulting a pillar taper of 0 (θ =88°); (b) with reflowed photoresist and 10 sccm of O₂, resulting a pillar taper of 0.8 (θ =51°); (c) with reflowed photoresist and 20 sccm of O₂, resulting a pillar taper of 1.4 (θ =35°); (d) with reflowed photoresist and 25 sccm of O₂, resulting a pillar taper of 1.2 (θ =40°).

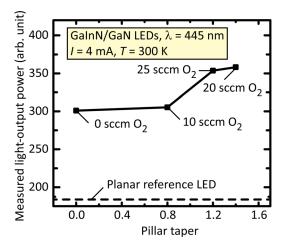


FIG. 4. Measured light-output power as a function of the pillar taper for a GaInN LED patterned with an array of TiO₂ micro-pillars. The TiO₂ micro-pillars have the same structure as described in Fig. 2. For comparison, a planar reference LED is also shown.

patterned with TiO₂ micro-pillars having various pillar tapers are fabricated and measured. The TiO2 micro-pillars have the same structure as described in Fig. 2. Figure 4 shows the measured LOP as a function of the pillar taper. As shown in Fig. 4, compared with planar reference LEDs, LEDs patterned with TiO₂ micro-pillars with vertical sidewalls show a 68% enhancement in LOP. And LEDs patterned with TiO₂ micro-pillars with tapered sidewalls show even higher LOP enhancement, with the LED patterned with TiO2 micropillars having a pillar taper of 1.4 shows the largest LOP enhancement of 100%. As shown in Fig. 4, when compared with planar reference LEDs, LEDs patterned with TiO₂ micro-pillars with vertical sidewalls (taper = 0) can increase the light extraction. Moreover, LEDs patterned with TiO₂ micro-pillars with tapered sidewalls can further increase the light extraction from LEDs. And the LED patterned with TiO₂ micro-pillars having a pillar taper that is closest to the optimum pillar taper, as predicted by the ray-tracing simulation, shows the largest enhancement in LOP. This trend is in good agreement with the ray-tracing simulation results.

Figure 5(a) shows the measured far-field emission intensity as a function of the angle of emission for a LED patterned with TiO₂ micro-pillars with tapered sidewalls (taper = 1.4), a LED patterned with TiO_2 micro-pillars with vertical sidewalls (taper = 0), and a planar reference LED. As shown in Fig. 5(a), compared with the planar reference LED, which have a Lambertian emission pattern, the LED patterned with TiO₂ micro-pillars with vertical sidewalls shows higher light emission intensity along off-surfacenormal directions, and the LED patterned with TiO₂ micropillars with tapered sidewalls shows much higher light emission intensity from off-surface-normal directions. Since the light emission along off-surface-normal directions is an indication of the light extracted from the sidewalls of pillars, we can see that micro-pillars with tapered sidewalls effectively promote the light extraction for LEDs by out-coupling light rays from the tapered sidewalls, as predicted by the raytracing simulation results. Furthermore, the angle of the peak emission intensity, which is the angle at which the emission intensity is at its maximum, deviates more from the

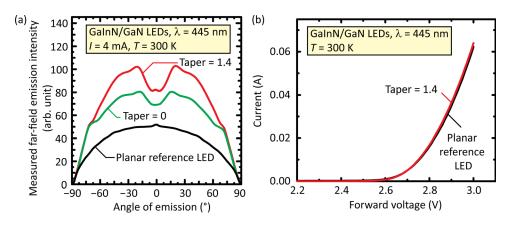


FIG. 5. (a) Measured far-field emission intensity as a function of the angle of emission for a LED patterned with TiO_2 micro-pillars with tapered sidewalls (taper = 1.4), a LED patterned with TiO_2 micro-pillars with vertical sidewalls (taper = 0), and a planar reference LED. (b) Current–voltage characteristic for a LED patterned with TiO_2 micro-pillars with tapered sidewalls (taper = 1.4) and a planar reference LED.

LED-surface-normal (i.e., a larger off-surface-normal angle) for the LED patterned with TiO_2 micro-pillars with tapered sidewalls than the LED patterned with TiO_2 micro-pillars with vertical sidewalls. This is another indication of the more effective light extraction from micro-pillars with tapered sidewalls, since the extracted light rays from tapered sidewalls primarily contribute to the emission along off-surface-normal directions. Figure 5(b) shows the current-voltage characteristic for a LED patterned with TiO_2 micro-pillars with tapered sidewalls (taper = 1.4) and a planar reference LED. As shown in Fig. 5(b), the current-voltage characteristic remains the same for both samples, indicating that the active region as well as the contacts are not damaged during the dry etch.

In summary, we have demonstrated an effective and elegant method to enhance the light extraction for GaInN LEDs. The method employs TiO₂ micro-pillars with tapered sidewalls that are refractive-index-matched to the underlying GaN layer. The tapered micro-pillars are fabricated by using reflowed photoresist as mask during CHF₃-based dry etch, with O₂ added in order to precisely control the taper angle. LEDs patterned with TiO₂ micro-pillars having a pillar taper of about 1.4 show a 100% enhancement in LOP over planar reference LEDs. The measured results are in good agreement with the ray-tracing simulation results, showing the strong potential of optical surfaces that are controlled in terms of refractive index and lateral structure to attain high light extraction in GaN-based LEDs.

The authors gratefully acknowledge support by Samsung LED, Korean Ministry of Knowledge Economy and Korea Institute for Advancement of Technology through International Collaborative R&D Program, the National Science

Foundation, Sandia National Laboratories, Department of Energy, Magnolia Optical Technologies, and Raydex Technology, Inc. Authors J.C. and E.F.S. were supported by Sandia's Solid-State Lighting Sciences Center, an Energy Frontier Research Center funded by the U.S. Department of Energy, Office of Basic Energy Sciences.

¹M. R. Krames, O. B. Shchekin, R. Mueller-Mach, G. O. Mueller, L. Zhou, G. Harbers, and M. G. Craford, J. Disp. Technol. 3, 160 (2007).

²E. F. Schubert, *Light Emitting Diodes*, 2nd ed. (Cambridge University Press, Cambridge, 2006).

³W. C. Peng and Y. C. S. Wu, Appl. Phys. Lett. **89**, 041116 (2006).

⁴T. Fujii, Y. Gao, R. Sharma, E. L. Hu, S. P. Denbaars, and S. Nakamura, Appl. Phys. Lett. **84**, 855 (2004).

⁵J. J. Wierer, Jr., A. David, and M. M. Megens, Nat. Photonics 3, 163 (2009).

⁶D. H. Kim, C. O. Cho, Y. G. Roh, H. Jeon, Y. S. Park, J. Cho, J. S. Lm, C. Sone, Y. Park, W. J. Choi, and Q. H. Park, Appl. Phys. Lett. 87, 203508 (2005).

⁷J. Cho, H. Kim, H. Kim, J. W. Lee, S. Yoon, C. Sone, Y. Park, and E. Yoon, Phys. Status Solidi C 2, 2874 (2005).

⁸H. Gao, F. Yan, Y. Zhang, J. Li, Y. Zeng, and G. Wang, J. Appl. Phys. 103, 014314 (2008).

⁹A. N. Noemaun, F. W. Mont, J. Cho, E. F. Schubert, G. B. Kim, and C. Sone, J. Vac. Sci. Technol. A 29, 051302 (2011).

¹⁰A. N. Noemaun, F. W. Mont, G. B. Lin, J. Cho, E. F. Schubert, G. B. Kim, C. Sone, and J. K. Kim, J. Appl. Phys. **110**, 054510 (2011).

¹¹D. A. Stocker, E. F. Schubert, and J. M. Redwing, Appl. Phys. Lett. **73**, 2654 (1998).

¹²C. F. Lin, J. J. Dai, Z. J. Yang, J. H. Zheng, and S. Y. Chang, Electrochem. Solid-State Lett. 8, C185 (2005).

¹³J. Q. Xi, M. F. Schubert, J. K. Kim, E. F. Schubert, M. Chen, S. Y. Lin, W. Liu, and J. A. Smart, Nat. Photonics 1, 176 (2007).

¹⁴M. K. Lee, C. L. Ho, and C. H. Fan, Appl. Phys. Lett. **92**, 061103 (2008).

¹⁵O. B. Shchekin, J. E. Epler, T. A. Trottier, T. Margalith, D. A. Steigerwald, M. O. Holcomb, P. S. Martin, and M. R. Krames, Appl. Phys. Lett. 89, 071109 (2006).