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Enhanced light extraction in n-GaN-based light-emitting diodes with three-dimensional semi-spherical structure

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Three-dimensional (3D) periodic micro/nanostructures can have a remarkable enhancement effect on light-emitting diodes (LEDs). However, simple, high-throughput and large-area fabrication of 3D periodic micro/nanostructures with a high duty ratio is difficult. In this Letter, high-duty-ratio 3D semi-spherical structures were fabricated on the surface of n-GaN-based vertical-structure LEDs by under-exposure ultraviolet lithography and dry etching. The resulting LEDs provide about 200% more light output power than those with a flat surface. This method of fabricating high-duty-ratio 3D semi-spherical structures could be used in other optical devices and shows potential for industrial production and commercialization. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4865417]

GaN-based light-emitting diodes (LEDs) have been developed substantially in past decades because of their variety of applications, including backlighting in liquid crystal displays, traffic signal lamps, and general illumination.[1–3] The efficient extraction of light from LEDs is a focus of attention in the drive to realize LEDs with high external quantum efficiency and is currently a bottleneck restricting their application. There are two principal approaches to improve external quantum efficiency η_{ext} ($\eta_{ext} = \eta_{in} \times \eta_{extraction}$): the first to increase internal quantum efficiency (η_{in}), and the second to maximize light extraction efficiency ($\eta_{extraction}$). The best way to improve the luminous efficiency of LEDs is to increase their $\eta_{extraction}$ because further improvement of η_{in} may not be readily achieved given its value has reached nearly 80%.[4,5] Light can only travel from the GaN layer to air within a critical angle of 23.6° because of the large difference in refractive index between a GaN film (n = 2.5) and air (n = 1.0). Roughening the top surface of LEDs is an effective method to improve light extraction because it can reduce internal light reflection and scatter the light outward. Various surface roughening structures, such as cone-like surfaces,[6–8] honeycomb structures,[9] and microlens arrays,[10] have been obtained *via* dry or wet etching techniques.

Theoretical calculations[11] have revealed that three-dimensional (3D) periodic structures can effectively enhance light extraction efficiency, especially hexagonal and semi-spherical structures. Hexagonal cone-like structures have been obtained by photoelectrochemical wet etching,[8] but non-uniformity and a large leakage current are associated with wet etching. An array of semi-spherical structures was formed on the surface of n-GaN-based vertical LEDs (V-LEDs) by a photoresist reflowing method after ultraviolet (UV) lithography.[10] However, the low duty ratio of the resulting microlens array is a disadvantage of this method, so the enhancement of light output power is limited. Meanwhile, low throughput or high cost is associated with current techniques to fabricate complicated 3D periodic structures, such as direct laser writing,[12,13] laser interference lithography,[14] and gray exposure,[15,16] which limits large-scale industrial production. Hence, inexpensive, large-scale, and industry-compatible methods to enhance the light extraction efficiency of LEDs are needed to facilitate industrial and commercial use. In this paper, 3D semi-spherical structures with a high duty ratio (about 100%) are fabricated by an under-exposure method based on UV lithography and dry etching. These structures can dramatically improve the light extraction efficiency of n-GaN-based V-LEDs.

In this study, n-GaN-based V-LEDs with a top layer of n-GaN were fabricated by a laser lift-off (LLO) process.[8,17,18] First, Ag and Au were successively deposited on the surface of p-type GaN. Then, the chip was flipped and bonded to an Au/Sn alloy-coated Si substrate. A KrF laser was used to decompose the GaN and separate the chip from the sapphire substrate. After the LLO process, the sample was thinned. This was followed by n-type electrode deposition; the thickness of the n-GaN layer was about 8 μm. A 1.2-μm-thick layer of positive photoresist S1813 (Shipley microposit S1813, Massachusetts, USA) was spin coated onto the substrate and pre-baked on a hot plate at 115 °C for 60 s. The photoresist patterns were exposed using a UV mask aligner (Karl Süss MA6) in hard-contact exposure mode. The light source was a mercury lamp with a wavelength of 365 nm. The photoresist patterns were transferred onto the n-GaN layer by inductively coupled plasma reactive ion etching (ICP-RIE; PlasmaLab System 100, Oxford Instruments) with a mixture of BCl_3 and Cl_3 gases. A focused ion beam system (FIB DB235, FEI Company, USA) was used to study the surface morphology of the LEDs. Electroluminescent (EL) spectra were obtained by an integrating sphere (Labsphere LMS 100, Labsphere Inc., USA), and current-voltage (*I-V*) characteristics were measured by a semiconductor characterization system (Keithley 4200 SCS, USA).

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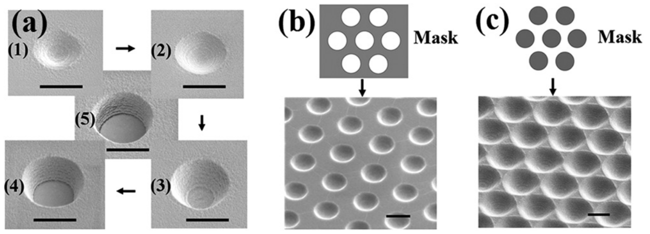

FIG. 1. (a) Scanning electron microscopy (SEM) images of photoresist layers following UV exposure at doses of (1) 30 mJ/cm^2, (2) 60 mJ/cm^2, (3) 90 mJ/cm^2, (4) 120 mJ/cm^2, and (5) 150 mJ/cm^2. (b) SEM image of the photoresist layer obtained using a dark-field mask. (c) SEM image of the photoresist layer obtained using a bright-field mask. The scale bar is 2 μm.

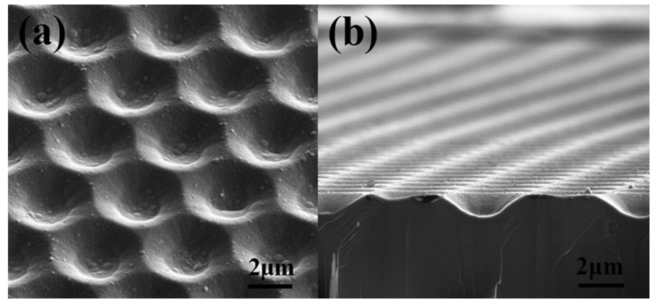

FIG. 2. (a) SEM image of a 3D semi-spherical structure with a pitch of 4 μm on the surface of a n-GaN-based V-LED. (b) Cross-sectional SEM image of the 3D semi-spherical structure.

Theoretical calculations revealed that the light intensity distribution can be changed from a parallel distribution to a Gaussian distribution[19] when the exposure dose decreases dramatically. We verified the theoretical calculation results using photoresist S1813, as shown in Figure 1(a). The diameter of the hole of the dark-field mask is 3 μm. In Figure 1(a), (1), (2), (3), (4), and (5) show scanning electron microscopy images of the photoresist pattern at exposure doses of 30, 60, 90, 120, and 150 mJ/cm^2, respectively. For the normal exposure dose (150 mJ/cm^2), the mask patterns were transferred onto the photoresist consistently. The profiles of the photoresist layers changed with the decreasing exposure dose. A semi-spherical structure was obtained when the exposure dose was about one third of the normal exposure dose. Therefore, using light with a Gaussian distribution intensity at under exposure, photoresist patterns with a 3D semi-spherical profile can be obtained, as shown in Figure 1(b). However, the semi-spherical structure array obtained using a dark-field mask has a low duty ratio because the diameter of the pit is less than the hole of the dark-field mask for the under-exposure method. In contrast, the complementary bright-field mask allows larger light transmittance than the corresponding dark-field mask, so 3D semi-spherical structures with a high duty ratio can be obtained using the complementary bright field mask at the same exposure, as shown in Figure 1(c). This difference will be analyzed further in another article. We found that the duty ratio of the semi-spherical structures reached almost 100% using the bright-field mask.

The photoresist pattern was transferred onto the n-GaN surface through an ICP dry-etching technique. To guarantee the equivalent photoresist shape was transferred onto n-GaN, we used an etching ratio of photoresist to n-GaN of about 1. Large areas on the surface of n-GaN-based V-LEDs were covered with uniform 3D semi-spherical structures with a pitch of 4 μm using this approach, as shown in Figure 2. The depth of each semi-spherical structure is about 1040 nm and can be controlled by the etching ratio between n-GaN and photoresist.

Light emitted from multiple quantum wells can only travel from the GaN layer to air within a critical angle of 23.6° for GaN-based LEDs with flat surfaces. However, the semi-spherical structures can reduce the total reflection of light and thus increase the output efficiency of LEDs, as illustrated in Figure 3(a). Blue emission from n-GaN-based V-LEDs with flat and 3D semi-spherical-structured surfaces was investigated by optical microscopy, as shown in Figure 3(b). A plan-view optical micrograph image reveals that the semi-spherical structure can markedly enhance the light extraction of n-GaN-based V-LEDs by reducing the reflected light.

Figure 4(a) depicts the *I-V* characteristics of V-LEDs with a flat surface and covered with semi-spherical structures with a period of 4 μm. The forward voltage and reverse leakage currents are almost unchanged by dry etching with ICP. This result reveals that the etching process has negligible influence on the electrical properties of the LED chips. The light output power of the V-LEDs with flat and semi-spherical-structured surfaces was measured with an injection current in the range of 50–380 mA, as shown in Figure 4(b). The light output power of the V-LED chip with semi-spherical structures shows an obvious enhancement compared with that of the flat V-LED; an enhancement of about 200% was observed when the injection current was 350 mA.

Angle-resolved EL spectra of the flat and semi-spherical LEDs at an injection current of 350 mA and integration of 100 ms are presented in Figure 5(a). Emission patterns were measured from $\theta = 0°$ to 180°, and the distance between fiber and chip was fixed at 30 cm, which is much larger than the size of the measured V-LED chip (1.2 mm × 1.2 mm). It is

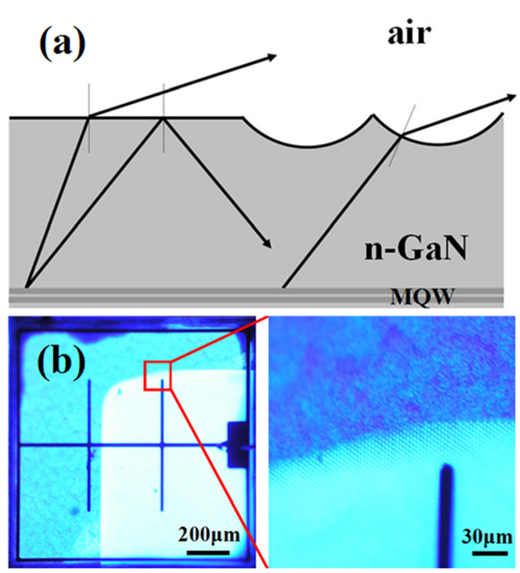

FIG. 3. (a) Diagram showing the refraction of light from a multiple quantum well (MQW) over flat and semi-spherical structures on the surface of n-GaN. (b) Plan-view optical micrograph of the light emission image of LED chips with flat and 3D semi-spherical-structured surfaces.

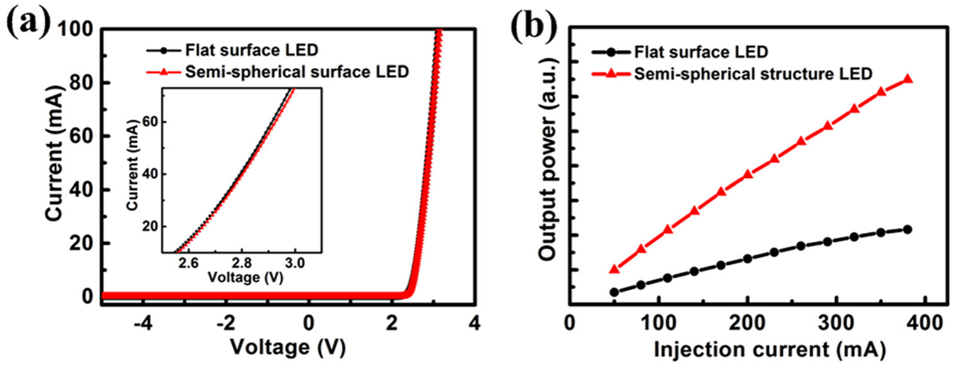

FIG. 4. (a) *I-V* characteristics of V-LEDs with flat and semi-spherical-structured surfaces (inset: enlarged view of *I-V* curves around 40 mA). (b) Light output power of V-LEDs with flat and semi-spherical-structured surfaces as a function of injection current.

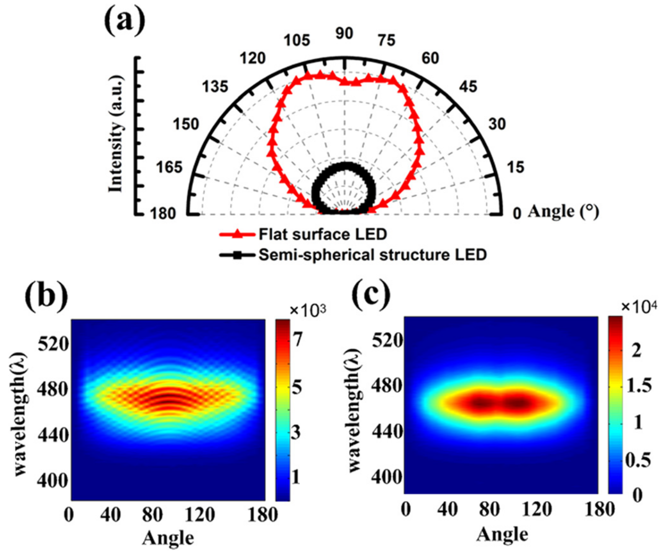

FIG. 5. (a) Angle-resolved EL spectra of V-LEDs with flat and semi-spherical-structured surfaces at an injection current of 350 mA. (b) Angular-resolved EL pattern from a flat-surfaced LED. (c) Angular-resolved EL pattern from a semi-spherical-structured LED.

evident that the emission intensities of the LED chip with semi-spherical structures are greatly enhanced in all directions compared with those of the flat LED. The largest intensity enhancement is obtained at 35–75° and 105–145°. Figures 5(b) and 5(c) show the angular emission patterns of the LEDs with flat and semi-spherical-structured surfaces. Large differences are observed between the patterns of these two types of LEDs. Multiple maxima caused by the Fabry-Perot effect are observed from the flat LED (Figure 5(b)). However, much of the light is directed outward rather than undergoing multiple reflections within GaN for the semi-spherical-structured LED, which shows a single broad peak (Figure 5(c)). The single broad peak suggests that the semi-spherical-structured surface allows randomization of light rays and diminishes the Fabry-Perot effect.

In summary, we improved the light extraction efficiency of V-LEDs by fabricating 3D semi-spherical structures with a high duty ratio on their surface using under-exposure UV lithography and dry etching. The light output power of V-LEDs with semi-spherical structures was enhanced dramatically compared with that of a flat V-LED. The method used to fabricate these high-duty-ratio 3D semi-spherical structures has the advantages of simplicity, large-area capability, high throughput, and low cost and could be used in other optical devices, showing potential for industrial production and commercialization.

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[1]E. F. Schubert and J. K. Kim, Science **308**, 1274 (2005).
[2]H. X. Jiang, S. X. Jin, J. Li, J. Shakya, and J. Y. Lin, Appl. Phys. Lett. **78**, 1303 (2001).
[3]O. 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).
[4]A. Hangleiter, D. Fuhrmann, M. Grewe, F. Hitzel, G. Klewer, S. Lahmann, C. Netzel, N. Riedel, and U. Rossow, Phys. Status Solidi A **201**, 2808 (2004).
[5]D. Fuhrmann, C. Netzel, U. Rossow, A. Hangleiter, G. Ade, and P. Hinze, Appl. Phys. Lett. **88**, 071105 (2006).
[6]J. H. Son, J. U. Kim, Y. H. Song, B. J. Kim, C. J. Ryu, and J.-L. Lee, Adv. Mater. **24**, 2259 (2012).
[7]H.-M. An, J. I. Sim, K. S. Shin, Y. M. Sung, and T. G. Kim, J. Quantum Electron. **48**, 891 (2012).
[8]T. Fujii, Y. Gao, R. Sharma, E. L. Hu, S. P. DenBaars, and S. Nakamura, Appl. Phys. Lett. **84**, 855 (2004).
[9]C. L. Lin, P. H. Chen, C.-H. Chan, C. C. Lee, C.-C. Chen, J.-Y. Chang, and C. Y. Liu, Appl. Phys. Lett. **90**, 242106 (2007).
[10]D. W. Kim, H. Y. Lee, N. Y. Cho, Y. J. Sung, and G. Y. Yeom, Jpn. J. Appl. Phys. **44**, L18 (2005).
[11]Q. H. Zheng, B. L. Liu, and B. P. Zhang, Chin. J. Electron Devices **31**(4), 1077 (2008).
[12]S. Tottori, L. Zhang, F. M. Qiu, K. K. Krawczyk, A. Franco-Obregón, and B. J. Nelson, Adv. Mater. **24**, 811 (2012).
[13]J. K. Gansel, M. Thiel, M. S. Rill, M. Decker, K. Bade, V. Saile, G. V. Freymann, S. Linden, and M. Wegener, Science **325**, 1513 (2009).
[14]N. D. Lai, W. P. Liang, J. H. Lin, C. C. Hsu, and C. H. Lin, Opt. Express **13**, 9605 (2005).
[15]J. H. Lake, S. D. Cambron, K. M. Walsh, and S. McNamara, IEEE J. Microelectromech. Syst. **20**(6), 1483 (2011).
[16]L. Mosher, C. M. Waits, B. Morgan, and R. Ghodssi, IEEE J. Microelectromech. Syst. **18**(2), 308 (2009).
[17]H. W. Jang, S. W. Ryu, H. K. Yu, S. Lee, and J.-L. Lee, Nanotechnology **21**, 025203 (2010).
[18]S. J. Wang, K. M. Uang, S. L. Chen, Y. C. Yang, S. C. Chang, T. M. Chen, and C. H. Chen, Appl. Phys. Lett. **87**, 011111 (2005).
[19]P. Duan, C. Q. Ji, and X. D. Yuan, J. Dalian University **30**(3), 68 (2009).