Fabrication of large-scale spherical-cap structure and the application on n-GaN based light-emitting diodes

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An under exposure method based on Fresnel diffraction effect in a conventional optical lithography system is used to fabricate large-scale, uniform spherical-cap structures. This method provides an effective roughening technology on the top surface of light-emitting diodes (LEDs) to improve the light extraction efficiency of LEDs. LEDs with high duty cycle spherical-cap structures showed enhanced light output power by 130%–160% compared with the LED with a flat surface. This simple and easy shape control method has potential applications in other optical devices such as organic LEDs, inorganic solar cells, and laser diodes. © *2014 American Vacuum Society*.
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I. INTRODUCTION

GaN-based light-emitting diodes (LEDs) have recently attracted significant attention for their diverse applications, such as backlighting in liquid crystal displays, traffic signal lamps, vehicle lamps, and becoming a major contender in ecofriendly light sources.[1–3] The n-side-up GaN based vertical LEDs (V-LEDs) fabricated by laser lift-off (LLO) processes have been demonstrated to be very effective for high power operation.[4,5] Despite significant progress in recent years, the external quantum efficiency of GaN-based LEDs is still not high enough to realize LED-based solid state lighting. The external quantum efficiency is mainly limited by low light extraction efficiency. One of the primary reasons for low light extraction efficiency is total internal reflection at the interface due to the large difference in refractive index between the GaN film ($n = 2.5$) and air ($n = 1.0$). So the light can only travel from the GaN layer to air within a critical angle of 23.6°. Most of photons were trapped inside the GaN-based LED device and converted to heat, which degrades the performance and the durability of the device. Roughening the top surface of the LEDs is an effective and simple method to improve the light extraction, such as conelike surface,[6–8] honeycomb structure,[9] microlens arrays,[10–12] etc.

Theoretical calculation[13] presents that three dimension (3D) periodic structures show an extremely obvious enhancement effect on the LEDs light extraction efficiency. Meanwhile, the low throughput or high cost are associated with the techniques such as the direct laser writing,[14,15] the laser interference lithography,[16] and gray exposure,[17,18] which limits the large-scale industrial production. As we know, the large area and uniform microstructures can be obtained by ultraviolet (UV) or extreme ultraviolet (EUV) lithography with low cost and high throughput. However, most of these structures have a very simple two-dimensional geometry. Some 3D structures can be obtained by improving UV lithography, such as one step UV lithography,[19] inclined/rotated UV lithography,[20] microstereolithography,[21] moving mask UV lithography,[22] etc. But the duty-cycle of 3D structure fabricated by these methods is limited. In this study, we developed an approach to fabricate the 3D concave spherical-cap structures with high duty-cycle (about 100%) by exposure method and transfer to many kinds of substrates by dry etching. The GaN based V-LED chips with high duty-cycle concave spherical-cap structure show higher light output power than that of the LEDs with flat surface.

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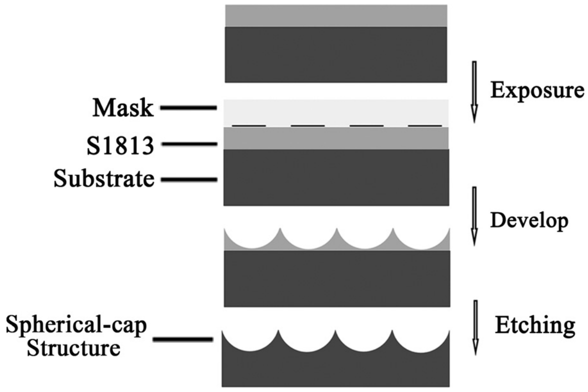

FIG. 1. Schematic diagram of the fabrication process.

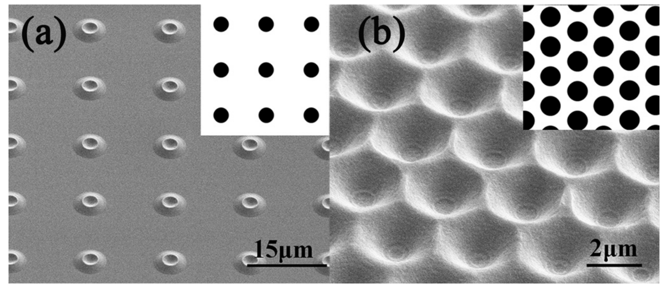

FIG. 2. SEM images of the resulting 3D photoresist structures fabricated by under exposure method. The exposure dose is (a) 110 mJ/cm^2 and (b) 50 mJ/cm^2. The inserts are the corresponding mask and the disk diameter, and the periods are 5 μm/15 μm (a) and 2 μm/3 μm (b), respectively.

II. EXPERIMENT

The schematic diagram of the fabrication process is shown in Fig. 1. First, 1.2-μm-thick layer of S1813 positive photoresist (Shipley microposit S1813, Massachusetts, USA) was spin-coated on the substrates (Si, quartz, GaN and GaN based V-LED) and then the samples were prebaked on a hotplate at 115 °C for 60 s. Second, the substrates were exposed using an ultraviolet mask aligner (Karl Süss MA6). The employed exposure mode was hard contact and the light source was a mercury lamp with a light wavelength of 365 nm. After exposure, the samples were developed in MF-319 developer for 40 s and rinsed with deionized water for 10 s. Finally, the photoresist patterns were transferred onto substrates by dry etching. In order to transfer the equivalent photoresist shape onto the substrates, the appropriate etching process with the etching rate ratio of photoresist to substrate of about 1 was selected.

In this work, the photoresist patterns were transferred into Si and quartz by reactive ion etching (RIE) system (PlasmaLab 80 Plus, Oxford Instruments) and transferred into the GaN and GaN based V-LED layer by inductively coupled plasma reactive ion etching (ICP-RIE) system (PlasmaLab System 100, Oxford Instruments). The light output power was obtained by an integrating sphere (Labsphere LMS 100, Labsphere Inc., USA); current–voltage (*I*–*V*) characteristics were measured by a semiconductor characterization system (Keithley 4200 SCS, USA).

III. RESULTS AND DISCUSSION

In optics, an Arago spot, Fresnel bright spot, or Poisson spot is a bright point that appears at the center of a circular object's shadow due to Fresnel diffraction which played an important role in the discovery of the wave nature of light. Fresnel diffraction lithography as a UV exposure technology has been reported.[23–26] Theoretical calculation[27] revealed that the light intensity distribution can be changed from a parallel distribution to a Gaussian distribution; this effect on photoresist will be more obvious in under exposure condition. The photoresist pattern is shown in Fig. 2(a), which is obtained at the exposure dose of 110 mJ/cm^2, lower than the normal exposure dose of 150 mJ/cm^2, and the corresponding mask disc is 5 μm diameter and 15 μm period with square mask pattern. We can find that the side wall of the obtained structure caused by Poisson spot is not sheer but inclined; we call this structure in the middle of the obtained pattern as a spherical-cap structure. The radius of the Poisson spot will get bigger when decreasing disk diameter (with fixed other parameters).[25] So if we reduce the disc diameter of the mask, the diameter of the Poisson spot will be increased, and then, the diameter of the spherical-cap structure will be also increased. The spherical-cap structure will connect each other when there is a decrease in the period of the disc, and the high duty-cycle spherical-cap structure can be obtained. Figure 2(b) shows the high duty-cycle 3D spherical-cap structure array using the mask disc of 2 μm diameter and 3 μm period with hexagonal arrangement, and the exposure dose is 50 mJ/cm^2. The uniform 3D spherical-cap photoresist

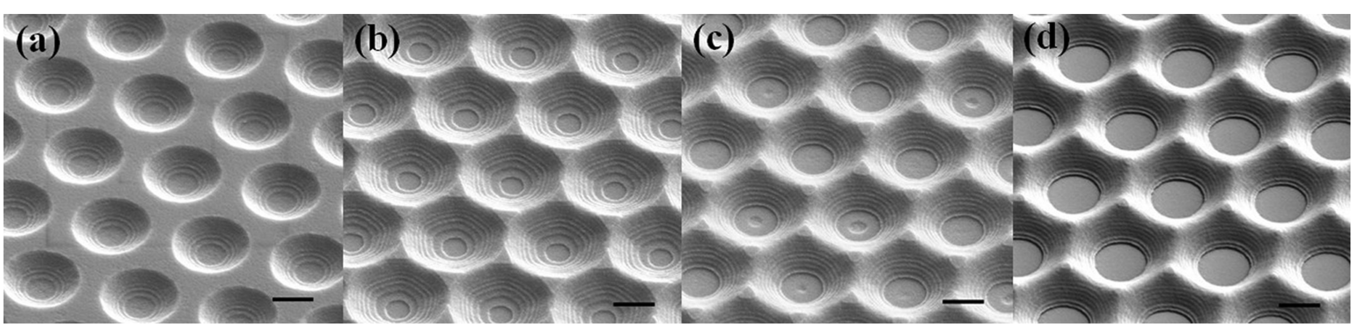

FIG. 3. SEM image of the 3D spherical-cap resist structure with a period of 5 μm at the exposure dose of (a) 48, (b) 56, (c) 64, and (d) 72 mJ/cm^2, respectively. The scale bar is 2 μm.

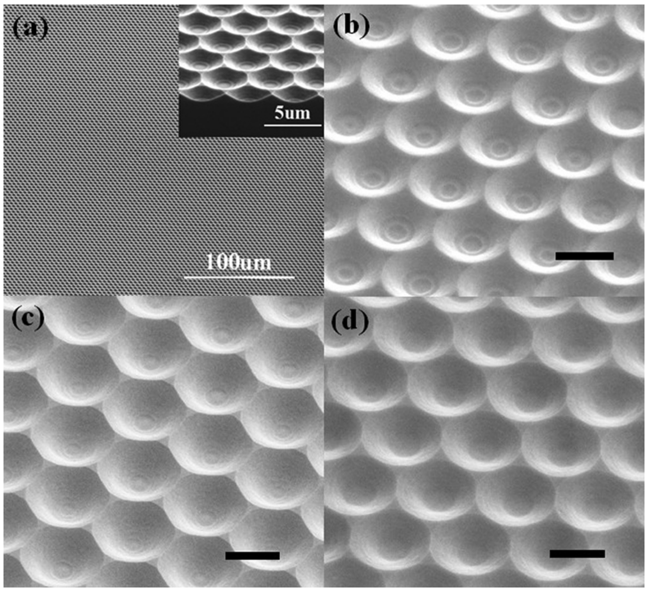

FIG. 4. (a) SEM image of the large area of the 3D spherical-cap structure. The insert shows the cross-sectional SEM image of the spherical-cap structure. The SEM image of the 3D spherical-cap structure on (b) Si, (c) quartz, and (d) n-GaN, the exposure parameters are the same. Si and quartz are etched by RIE; n-GaN is etched by ICP. The scale bar is 2 μm.

structure with large area can be obtained by under exposure method based on Fresnel diffraction effect in single step.

Besides the mask type, the exposure dose can also affect the obtained structure morphology. Figure 3 shows the SEM images of the obtained spherical-cap resist structure at various exposure doses, and the mask disc with 3 μm diameter and 5 μm period. We can find that the diameter and the height of the spherical-cap increase with increasing exposure dose; the perfect high duty-cycle spherical-cap structure can be obtained at the exposure dose about 56 mJ/cm^2. And by further increasing the exposure dose, the exposure depth exceeds the thickness of the photoresist (1.2 μm), so the bottom of the spherical-cap structure becomes flat.

The large-area with uniform and high duty-cycle 3D spherical-cap photoresist structure can be obtained through optimizing the mask type and exposure dose, and the perfect spherical-cap structure can be seen from the cross-sectional image, as shown in Fig. 4(a). By controlling the dry etching parameters, the spherical-cap structure can be transferred into kinds of substrates. Figures 4(b)–4(d) show the transferred results on the Si, quartz, and GaN substrate, respectively. We can find that the large area 3D concave spherical-cap structure with high duty-cycle on many type substrates can be obtained by using the under exposure method and dry etching process, which greatly expands the range of applications.

In this study, n-GaN-based V-LEDs with a top layer of n-GaN were fabricated by LLO process.[4,5,8] 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.

The high duty-cycle 3D spherical-cap structures with periods of 3, 4, and 5 μm on n-GaN based V-LEDs surface were fabricated using the under exposure method and ICP dry etching, and the etching depth is about 1 μm. The *I–V* characteristics and light output power for the flat surface sample and the patterned samples were studied. Figure 5(a) depicts the *I–V* characteristics of V-LEDs with a flat surface and covered with spherical-cap structures with periods of 3, 4, and 5 μm. We can find that the forward voltage and the reverse leakage currents are almost the same for the entire sample after dry etching by ICP. The inset clearly shows that the *I–V* characteristics of the patterned LEDs near the threshold voltage exhibited almost the same *I–V* characteristics of the flat surface LEDs. The result reveals that the dry etching process has negligible influence on the electrical properties of the LED chips. The light output power of V-LEDs for the flat surface and the spherical-cap structure chips was measured with an injection current in the range of 50–380 mA at a wavelength of 470 nm, as shown in Fig. 5(b). The light output power of the V-LEDs with spherical-cap structures show a large enhancement compared with that of flat surface V-LED; an enhancement of about 140%, 160%, and 130% for the period of 3, 4, and 5 μm were observed when the injection current was 350 mA, respectively.

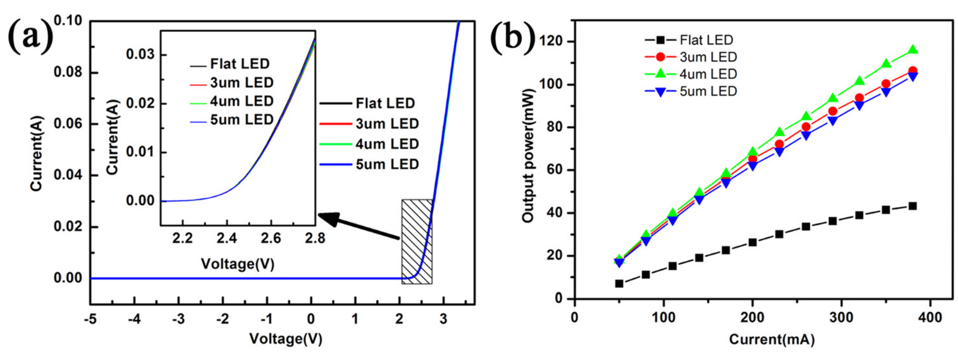

FIG. 5. (Color online) (a) Current–voltage (*I–V*) characteristics of V-LEDs for the flat surface sample and the spherical-cap structure samples with periods of 3, 4, and 5 μm (inset: enlarge view of *I–V* curves near the threshold voltage). (b)The light output power of V-LEDs with flat surfaces, the spherical-cap structure samples as a function of injection current (50 to 380 mA).

IV. CONCLUSION

In summary, we explored an under exposure method based UV lithography using the Fresnel diffraction effect. By this method, the high duty-cycle concave spherical-cap structures were fabricated on photoresist by changing the exposure dose and the pattern of the mask. After dry etching process, large-scale, uniform, and high duty-cycle spherical-cap structures with periods of 3, 4, and 5 μm were obtained. The light output power of V-LEDs with semi-spherical structures was enhanced greatly compared with that of the flat surface V-LED. This fabrication method has the advantages of large-area capability, low cost, and high throughput and could be used in other optical devices and provide a new method for industrial production and commercialization.

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