Fabrication of SiNₓ-based photonic crystals on GaN-based LED devices with patterned sapphire substrate by nanoimprint lithography

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Abstract: SiNₓ-based photonic crystal (PhC) patterns were fabricated on the ITO electrode layer of a GaN-based light-emitting diode (LED) device on a patterned sapphire substrate (PSS) by a UV nanoimprint lithography process in order to improve the light extraction of the device. A three-dimensional finite-difference time-domain simulation confirmed that the light extraction of a GaN LED structure on a PSS is enhanced when SiNₓ PhC patterns are formed on the ITO top layer. From the I-V characteristics, the electrical properties of patterned LED devices with SiNₓ-based PhC were not degraded compared to the unpatterned LED device, since plasma etching of the p-GaN or the ITO layers was not involved in the patterning process. Additionally, the patterned LED devices with SiNₓ-based PhCs showed 19%-increased electroluminescence intensity compared with the unpatterned LED device at 445 nm wavelength when a 20 mA current is driven.

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1. 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 general illumination both indoors and outdoors. However, 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 between a LED device and air, which is originated from the large difference in refractive index between GaN and air. By Snell’s law, the critical angle for a photon to escape from the device into air is about 24°, thus, a photon which propagates at an angle greater than the critical angle is guided and trapped inside the GaN-based LED device and is converted to heat, which degrades the performance and the durability of the device. Therefore, the enhancement of the light extraction is a crucial issue in improving the external quantum efficiency. To enhance the light extraction efficiency of GaN-based LEDs, various approaches, including the use of photonic crystals with large index contrast [1–3], microlens arrays [4–6], and self-assembled patterning [7] have been intensively conducted. The approach to address light extraction issue remains very important for achieving large external quantum efficiency in GaN-based LEDs. In addition to the light extraction issue, it is important to note that achieving high internal quantum efficiency is also important for realizing InGaN-based LEDs with high external quantum efficiency. The charge separation issue has been an important limiting factor in achieving high internal quantum efficiency in InGaN quantum well (QW) LEDs [8–14], especially for longer wavelength emission and high operating current density. Recent works for achieving high internal quantum efficiency in InGaN QW LEDs by charge separation suppression include semi/non-polar InGaN QW [8,9], c-plane InGaN QW with large overlap design [10–12], surface plasmon coupled InGaN QWs [13,14] approaches.

Especially, further enhancement of the external quantum efficiency of GaN-based LEDs on patterned sapphire substrates (PSSs) is greatly required. Most high quality GaN-based LEDs are fabricated on PSS in the current LED industry since the threading dislocation density in the epitaxial GaN layer is effectively reduced by the epitaxial lateral overgrowth and the micron-scaled patterns on the PSS act as scattering centers for the guided light inside LED devices [15–19]. The use of PSSs with micron-sized dimensions has led to increased light extraction efficiency in GaN-based LEDs. Recently, Lin and associates had demonstrated that there exists a dependency on the pattern coverage density on extraction efficiency enhancement for GaN LEDs grown on the micron-sized PSSs [20]. However, it is important to note that recent works by using nanoscale PSSs had also led to increase in internal quantum efficiency and light extraction efficiency in InGaN-based LEDs [21–24], as a result of two order-of-magnitude reduction in threading dislocation [21], reduction in screw dislocation density [22], and increase in light scattering by nano-scaled pattern [23,24]. Although both the internal quantum efficiency and light extraction efficiency of GaN-based LEDs are enhanced by the PSS, a further increase of the light extraction is essential for the realization of high brightness and high efficiency LEDs.

In this study, SiNx-based photonic crystal (PhC) structures are formed on the GaN-based LEDs, fabricated on the PSS, in order to increase the external quantum efficiency by nanoinprint lithography (NIL) [25–29], which offers low cost and high throughput compared to other lithography techniques such as photolithography [30], e-beam lithography [31] and so on. Prior to the NIL process, we analyzed the effect of the presence of the SiNx-based PhC on the light extraction of a GaN LED structure on a PSS using a three-dimensional finite-difference time-domain (FDTD) simulation tool. In addition, the optical and electrical properties of the patterned LED devices with SiNx-based PhC were confirmed by the electroluminescence (EL) and the I-V characteristics.
2. Experimental details

A typical GaN-based blue LED structure was grown on a (0001)-oriented PSS by a conventional MOCVD process. After the deposition of a thin low-temperature GaN buffer layer, the LED structure, which consists of layers of 5 µm thick un-doped GaN, 3 µm thick n-GaN, 50 nm thick InGaN/GaN multi QWs and 150 nm thick p-GaN, was fabricated. Then, a 200 nm thick ITO layer was sputtered onto the p-GaN layer to achieve current spreading between the p-pad metal and the p-GaN layer.

![Image of LED fabrication process]

Figure 1 shows the overall patterning process for the fabrication of the SiN$_x$/SiO$_2$ based PhC pattern on the LED structure. First, SiO$_2$/SiN$_x$ was deposited on the ITO layer by a PECVD process. The thickness of the SiO$_2$ layer was about 70 nm and the thickness of the SiN$_x$ layers was split between 300 nm and 500 nm in order to fabricate PhC patterns with different heights. After the PECVD process, a 200 nm thick LOL 2000™ sacrificial polymer layer was coated on the SiN$_x$ layer, followed by a UV NIL process at 20 atm of pressure while exposing the stack of the mold/resin/LED wafer to UV for 10 min. In the UV NIL process, a flexible polymer-based mold was used for conformal contact with the LED wafer [32]. A UV-curable
resin composed of 65 wt% of benzylmethacrylate base monomer, 5 wt % of Irgacure™ 184 UV initiator and 30 wt% of methacryloxypropyl-terminated polydimethylsiloxanes, was used for the UV NIL to elevate the etch resistance to the oxygen plasma [33]. After the UV NIL process, the sacrificial polymer layer under the imprinted pattern was cleared off with an oxygen plasma treatment. Next, a 50 nm thick Cr layer was deposited by e-beam evaporation and was lifted off the SiNₓ layer by removing the pattern, which was composed of the imprint resin/polymer sacrificial layer, with dimethylformamide solution. Finally, the masked SiNₓ layer with the Cr pattern was etched by a reactive ion etching process using CF₄ plasma and then the SiNₓ-based PhC patterns were formed on the LED structure.

To fabricate LED devices, at first, a photo-resist was coated onto the patterned LED wafer and was partially removed by photolithography to establish the contact region for p- and n-GaN. Prior to mesa etching, SiNₓ-based PhC patterns on the contact region for p- and n-GaN, which is not covered with the photo-resist, were removed along with the underlying SiO₂ layer by dipping the sample in buffered oxide etcher solution. Through mesa etching using ICP and deposition of p- and n-pad metals, composed of Cr/Au, 300 µm x 300 µm conventional lateral-type LED devices were fabricated. To analyze the optical and electrical properties of the patterned LED devices, measurements of EL and I-V characteristics were conducted. A three-dimensional FDTD simulation on light extraction of the patterned LED structures was carried out using a commercially available FullWAVE™ simulator [34–36].

3. Results and discussion

In Fig. 2, SiNₓ-based PhC patterns with heights of 300 nm and 500 nm, fabricated by the NIL and RIE processes, are shown. Figures 2(a) to 2(c) and Figs. 2(d) to 2(f) are SEM images of the cross-sectional, tilted and top views of PhC patterns with heights of 300 nm and 500 nm, respectively. Each well-aligned PhC pattern on the ITO electrode layer has a diameter of 250 nm and a pitch of 600 nm. Due to the difference of the etch resistance to CF₄ plasma between the SiO₂ and the SiNₓ layers, PhC patterns slightly display re-entrance etch-profiles. In this work, the etch rates of SiO₂ and SiNₓ layers, deposited by PECVD, were about 25 nm/min and 50 nm/min, respectively. This re-entrance profile is clearer in the PhC pattern of 500 nm in height than the PhC pattern of 300 nm in height. Figure 3(a) is the top SEM image of the fabricated LED device with the SiNₓ PhC pattern. The SiNₓ PhC pattern was only formed on the ITO top electrode layer of the LED device. Thus, there is no problem in forming the p- and n-pad metals on the p-GaN and n-GaN, respectively. Figure 3(b) is the cross-sectional SEM image of the GaN-based LED structure with the SiNₓ PhC pattern, which was grown on the PSS with a diameter of 2.5 µm and a height of 1.5 µm. As shown in Fig. 3(b), the array of SiNₓ PhC patterns was uniformly fabricated on the ITO electrode layer of the GaN-based LED.
Fig. 2. (a)–(c) are cross sectional, tilted and top views of SEM micrographs of 300 nm-high SiN, PhC patterns on the ITO electrode, respectively. (d)–(f) are also cross sectional, tilted and top views of SEM micrographs of 500 nm-high SiN, PhC patterns on the ITO electrode, respectively.

Fig. 3. (a) Top and (b) cross sectional SEM images of the LED device with the SiN, PhC pattern.

To investigate the effect of the presence of the SiN<sub>x</sub>-based PhC patterns with different heights on the light extraction of LED structures, a three-dimensional FDTD simulation was conducted. Figures 4(a) to 4(c) are schematic diagrams of the simplified LED structures for
the FDTD simulation, which are composed of a 200 nm thick ITO, a 9 µm thick GaN and 2 µm thick sapphire layers. The plane of continuous polarized dipoles was placed at 150 nm below the GaN surface as the light source which emits photons in random directions and the wavelength of the light source was set to 450 nm. The area of the simulation domain is limited to 3 µm x 5.1 µm and only the 300 nm-thick sapphire layer was partially involved in the simulation domain, in order to avoid the FDTD calculation being hugely time consuming. Three different LED structures, consisting of an LED on a flat sapphire substrate, an LED on PSS and an SiN_{x}-PhC patterned LED on PSS, were considered as shown in Figs. 4(a) to 4(c). The lens-shaped PSS pattern in the simulation structure has a 2 µm diameter, 3 µm pitch and 1 µm height and is pseudo-hexagonally arrayed. The SiN_{x}-PhC pattern in the simulation structure has a 300 nm diameter and 600 nm pitch and its height is split into levels from 100 nm to 900 nm with an increment of 200 nm. The grid size of the FDTD was set to 10 nm for reliable simulation computation, and periodic boundary conditions were applied to the x-y plane in order to minimize the effect of the small size of the simulation domain.

![Simulation Designs](image)

Fig. 4. Simplified FDTD simulation designs of (a) the conventional LED, (b) the LED with the PSS and (c) the LED with SiN_{x}-PhC patterns and the PSS.

Figure 5 presents the results of the FDTD simulation for the considered LED structures, which are described in Fig. 4. By inserting the PSS into the normal LED structure, the light extraction efficiency was increased by 18.75%. This agrees well with lots of reports that a PSS is helpful in enhancing the light extraction of LEDs, as well as improving the crystal quality of GaN by reducing the threading dislocation density. When the SiN_{x}-based PhC pattern with 300 nm of height is formed on the ITO top layer of the GaN LED structure, the light extraction efficiency was increased by up to 14.80%. Thus, from the simulation result, we confirmed further enhancement in the light extraction of a GaN-based LED on a PSS by introducing the additional patterned layer. This simulation method did not take into account the photon recycling and reabsorption process, thus this method may not provide the accurate absolute value of the light extraction efficiency. However, the use of this method is sufficient for providing comparison of the light extraction efficiency among all the LEDs.
Fig. 5. FDTD simulation results on light extraction of the conventional LED, the LED on the PSS and the SiNₓ-PhC patterned LED.

We measured the I-V characteristics of the unpatterned and SiNₓ-based PhC patterned LED devices, which were all fabricated on the PSS, as shown in Fig. 6. The forward voltages of all LED devices are in the 4.1 V to 4.2 V range at 20 mA drive current. The inset in Fig. 6 shows that the I-V characteristics of the patterned LEDs exhibited reduction in leakage current, in comparison to that of the unpatterned LED. This result can be explained by the presence of the thin SiO₂ layer on the ITO electrode, which acts as a surface passivation layer to prevent surface leakage [37, 38]. Thus, the electrical properties of the patterned LED devices with SiNₓ-based PhC were not degraded since no plasma etching process was conducted in the p-GaN layer while fabricating the SiNₓ-based PhC pattern on the ITO electrode.

Fig. 6. The I-V characteristics of the un-patterned LED device and LED devices with SiNₓ-PhC patterns of 300 nm and 500 nm in height. All LED devices were fabricated on the PSS. The inset shows the I-V characteristics on a logarithmic scale.

In order to confirm the effect of the presence of SiNₓ PhC pattern on the light extraction of the LED device, which was fabricated on the PSS, we measured the EL intensities of the
unpatterned LED device and the patterned LED devices with SiN$_x$ PhC patterns, as shown in Fig. 7. When 20 mA of current is injected, the EL intensities of the LED devices with SiN$_x$-based PhC patterns of 300 nm and 500 nm in height were increased by 14.5% and 19%, respectively, compared to that of the unpatterned LED device at a wavelength of 445 nm, as shown in Fig. 7(a). In contrast to the simulation results, the LED device with the 300 nm-high SiN$_x$-based PhC pattern showed stronger EL intensity than the LED device with the 500 nm-high SiN$_x$-based PhC pattern. The different tendencies shown by the EL and the simulation might result from slight differences in structure between the fabricated SiN$_x$-based PhC pattern and the designed SiN$_x$-PhC pattern in the simulation, including the diameter and profile. So far, most studies on the light extraction of LEDs have been performed on LEDs with flat sapphire substrates. Enhancing the light extraction of LEDs on PSS is relatively more difficult than it is for LEDs on flat sapphire substrates since the light extraction efficiency is already enhanced by the PSS. However, further increase in the light extraction of the LED device on the PSS was confirmed by forming the SiN$_x$-based PhC patterns, which suppress the total internal reflection. The EL intensity according to injection current at a wavelength of 445 nm is shown in Fig. 7(b). At every injection current, the patterned LED devices with SiN$_x$-based PhCs showed higher EL intensity than the unpatterned LED device.

![EL intensity at 20 mA current and EL intensity versus injection current at a wavelength of 445 nm for the un-patterned LED device and the patterned LED devices with SiN$_x$-based PhCs.](image)

Fig. 7. (a) EL intensity at 20 mA current and (b) EL intensity versus injection current at a wavelength of 445 nm for the un-patterned LED device and the patterned LED devices with SiN$_x$-based PhCs.
4. Conclusion

SiN$_x$-based PhC patterns were formed on GaN-based LED devices fabricated on PSS by the UV-NIL process in order to enhance the light extraction efficiency. From the three-dimensional FDTD simulation, when the height of the SiN$_x$-pattern is 300 nm, the light extraction efficiency of the SiN$_x$-PhC patterned LED structure on the PSS was increased by up to 14.80% compared to the unpatterned LED structure on the PSS. Similar to the simulation results, the SiN$_x$-based PhC patterned LED device on the PSS showed an increase in EL intensity of up to 19% compared to an unpatterned LED device on the PSS at 20 mA drive current. Thus, total internal reflection was suppressed inside the GaN-based LED device by inserting SiN$_x$-based PhC patterns. Moreover, the electrical properties of all patterned LED devices were not degraded because plasma etching of the p-GaN layer was not performed in the patterning process.

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