Fabrication of three-dimensional autocloned photonic crystal on sapphire substrate

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We applied the laser interference lithography method to form a patterned sapphire substrate (PSS). A three-dimensional photonic crystal was formed by autocloning the PSS with alternate Ta₂O₅/SiO₂ coatings. A high total integrated reflectance (TIR) band was obtained around the 410 to 470 nm wavelength range that matched the emission spectrum of the gallium nitride (GaN) light-emitting diode (LED) for application in manipulating the light extraction of the sapphire-based GaN LED. © 2010 Optical Society of America


1. Introduction

A three-dimensional (3D) autocloned photonic crystal (APhC) [1,2] is a multilayer structure with alternating high and low refractive index films stacked on a periodically corrugated substrate such that the third dimension periodicity is created on the two-dimensional periodic substrate. The deposition parameters of the films have to be properly controlled [3,4] such that the periodically corrugated substrate profile can be automatically replicated through the films. Depending on the application of the APhC, the substrate could be silicon for a photonic crystal waveguide [5,6], glass for a polarization selective grating [7], and wavelength filters [8]. It is important to note that the starting profile of the corrugated substrate has to be well patterned periodically. Numerous fabrication methods, such as photolithography, nanoimprint, and electron-beam writing [9] were used to form the submicrometer patterns on the substrate. It is noteworthy that the laser interference lithography (LIL) method is a low cost, maskless method for rapidly patterning the submicrometer array on the substrate [10–12]. APhC was applied on the sapphire substrate to enhance the light extraction efficiency of the gallium nitride (GaN) light-emitting diode (LED) that was laid on the other side of the sapphire [13]. It is important in this application that the sapphire substrate [2 in. (5 cm) diameter] should be patterned reliably and uniformly in a large area and that the periodicity of the APhC should be properly chosen such that the emission band of the GaN falls in the photonic bandgap of the APhC. In this paper, we report the details of patterning the sapphire substrate with the LIL method and the subsequent autocloning process for application in light extraction enhancement of a GaN LED.

2. Fabrication of the Two-Dimensional Patterned Sapphire Substrate with Laser Interference Lithography

A [100] orientation double-side-polished sapphire substrate with a 100 μm thickness of 2 in. (5 cm) diameter was first coated with a 250 nm thickness layer of SiNₓ by using the plasma-enhanced chemical vapor deposition method. The SiNₓ layer will be used as a hard mask for sapphire in the inductive coupled plasma (ICP) etching process. A 100 nm thick absorptive antireflection polymer film (ARP) was spin coated on the SiNₓ film for antireflection of the
325 nm He–Cd laser. A 300 nm thick photoresist [(PR) THMR-M100 by Tokyo Ohka Kogyo Co., Ltd.] was then spin coated on the ARP. The layer structure is shown in Fig. 2(a); it was then subjected to UV exposure by the LIL apparatus shown in Fig. 1. The UV source was a He–Cd laser operated at the 325 nm wavelength. The LIL was a single-beam system. The interference pattern and periodicity could be controlled by rotating the sample. In our LIL process, the exposures were performed in the PR with 37° angle of incidence for a constant pitch of 300 nm. The substrate was rotated 90° for a second exposure. After PR developing, as shown in Fig. 2(b) schematically and with the scanning electron micrograph (SEM) picture, the substrate was then subjected to reactive ion etching (RIE) to transfer the pattern to the SiN\textsubscript{x} layer. Inductive coupled plasma etching was then applied to transfer the pattern to the sapphire, and the remaining SiN\textsubscript{x} was removed by a dip in buffered oxide etchant. The resultant patterned sapphire substrate (PSS) is shown in Fig. 2(d) schematically and with the SEM picture. It can be seen that the pattern was in a rounded cone shape of \( \sim 110 \) nm in height and \( \sim 135 \) nm in diameter, and it was in a square-symmetrical lattice with a 300 nm side length. The process parameters of the RIE and ICP are listed in Table 1. The whole process is shown schematically in Figs. 2(a)–2(d) together with the corresponding SEM picture for the end product of each step.

In the LIL process, the laser exposure dose is crucial to the formation of the pattern on the PR. Although the PR that we used was a positive PR, nevertheless, the resulting pattern on the PR after the developing, could be changed from “positive” to “negative” as the laser dose increases. Initially, at a low exposure dose (110 mJ/cm\(^2\)), arrays of holes [dark hole as shown in the inset of Fig. 3(a)] were formed on the positive PR, the constructive interference peak located at the positions of the holes. As the laser dose was increased up to 200 mJ/cm\(^2\), the diameters of the holes increased due to the fact that the area over which the laser intensity was above the threshold increased and the neighboring holes started coalescing. Increasing the dose continuously, the saddle between the holes disappeared completely, leaving an isolated PR column over which the interference intensity was destructive. The diameters of the columns decreased as the dose further increased. The hole array transformed into a column array beyond a critical threshold, \( \sim 200–250 \) mJ/cm\(^2\) in the laser dose. Figure 3 shows the diameter of the hole and the column versus the laser dose. The insets of Figs. 3(a)–3(f) show the SEM pictures that revealed the evolution of the arrays pattern from holes to columns as the dose increases. The hole diameter varied from 250 to 260 nm, and the column diameter varied from 162 to 122 nm. The pattern changed from “positive” to “negative” as the laser dose increased over \( \sim 200–250 \) mJ/cm\(^2\). In our fabrication process for the APhC, we used the column array, Fig. 3(e), which was obtained by using a 275 mJ/cm\(^2\) laser dose.

### 3. Fabrication of Three-Dimensional Autocloned Photonic Crystal

The PSS was then subjected to the ion beam sputtering process. Ta\textsubscript{2}O\textsubscript{5} and SiO\textsubscript{2} films were coated

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<th>Table 1. Process Parameters for RIE and ICP Etching</th>
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\textsuperscript{a}SCCM denotes cubic centimeters per minute at standard temperature and pressure.
alternately on the patterned side of the PSS. In order to replicate the surface morphology of the multilayer films being deposited, it is important to provide steady and repeatable control of the coating parameters such as ion beam current and accelerating voltage, such that the corrugation of the PSS is continuously replicated. More importantly, because the autocloning process depends on the balance between the angular-dependent deposition rate and the angular-dependent etching rate [4], we applied RF bias on the substrate during the Ta$_2$O$_5$ deposition as the source for etching. The RF power was crucial to the success of the autocloning process, and we found that 30 W RF power in our apparatus produced the best autocloning effect. The coating parameters of the ion beam sputter process for Ta$_2$O$_5$ and SiO$_2$ layers were given in Ref. [4]. Figure 4(a) shows the schematics of the 3D photonic crystal; Figs. 4(b) and 5(c) show the SEM pictures of a typical 3D photonic crystal that we produced.

4. Optical Property of the Three-Dimensional APhC

We defined total integrated reflectance (TIR) as the sum of the intensities for the specular reflection and the diffractions to the front side of the APhC. The TIR can be readily measured by using an integrating sphere; it can also be simulated by using the finite-difference time-domain (FDTD) method. TIR is sensitive to the layer thickness. We simulated the TIR for different layer thicknesses; Figs. 5(a)–5(c) show the simulated TIR for Ta$_2$O$_5$ and SiO$_2$ thicknesses of 50, 60, and 70 nm each, respectively. In the
simulation, the refractive indices for the Ta$_2$O$_5$ and the SiO$_2$ layers were 2.2 and 1.46, respectively, the angle of incidence was 0°, there were 30 layers in total, the side length of the square lattice was 300 nm, and the thickness of the layers was defined as the tip-to-tip physical thickness. Figure 5(b) reveals that a high blue TIR band can be obtained around the GaN-based LED emission band (450–470 nm) for layer thicknesses of 60 nm each, and the TIR band varied drastically when the layer thicknesses changed. The solid curves in Figs. 6(a) and 6(b) show the measured TIR and the measured total integrated transmittance (TIT), respectively, for the fabricated APhC. The SEM picture revealed that the layer thicknesses were 58 nm each. The TIR and TIT that were simulated by using FDTD for an APhC with 58 nm layer thicknesses are shown as dashed curves in Figs. 6(a) and 6(b). The high TIR and the low TIT bands of the fabricated sample are close to that from the simulation. The 3D APhC fabricated by our process is, therefore, suitable for light extraction enhancement for the GaN-based LED.

5. Conclusion

A two-dimensionally PSS has been fabricated by using LIL and dry etching. By varying the exposure dose of the LIL, the patterned structure (hole or column and their diameters) can be controlled. The autocloning process of the alternate multilayer Ta$_2$O$_5$/SiO$_2$ was applied on the PSS to obtain the 3D APhC. The layer thicknesses of the 3D APhC were fine-tuned such that a high TIR band around the blue wavelength region was obtained. The structure is suitable for use as the substrate for the GaN-based LED to enhance the light extraction efficiency.

References