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Photonic crystal effect on light emission from InGaN/GaN multi-quantum-well structures

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Triangular hole arrays with nanoscaled lattice constants of 230 and 460 nm were fabricated on a *p*-type GaN epitaxial layer grown on an InGaN/GaN multi-quantum-well light emitting diode structure by metal-organic chemical vapor deposition. The hole geometries of dry-etched thin slabs for triangular lattice constants of 230 and 460 nm possessed diameters of 223 and 218 nm at the surface, and 108 and 76 nm at the bottom, with depths of 31 and 27 nm, respectively. The hole array with a lattice constant of 230 nm enhances photoluminescence intensity at wavelengths of 364 and 406 nm, but reduces light extraction at a wavelength of 450 nm, which indicates destructive surface diffraction correlated with light scattering in the photonic crystal structure. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2735927]

Solid-state lighting technology has recently advanced with the development of III-nitride semiconducting optoelectric devices and the implementation of nanophotonics.[1–3] The III-nitride optoelectronic devices have been investigated in both directions: both to increase the internal quantum efficiency by low dimensional structures such as quantum wells, quantum wires, and quantum dots in order to enhance the electron confinement for more photon emission,[4] and also to enhance the external quantum efficiency by introducing the nanophotonic structure of photonic crystals or nanoscaled surface roughness.[5] Nowadays, an internal quantum efficiency of up to 90% is reached, but the external quantum efficiency is less than 10%. The low light extraction is due to the relatively high refractive index of semiconductor materials in optoelectronic devices which cause internal scattering of light at the surface. Rather than being extracted, the photon is trapped inside by total internal reflection.[5,6] The application of photonic crystals for optoelectronic devices, especially light emitting diodes (LEDs), has been studied extensively.[7–9]

In this work, two dimensional photonic crystal (2D PC) slabs as periodic hole arrays were fabricated on the *p*-type GaN surface of an InGaN/GaN multi-quantum-well LED structure. Nanoscaled triangular lattice holes with lattice constants of 230 and 460 nm, for the diameter of 150 nm, were designed on a polymethylmethacrylate (PMMA) blocking film using an electron beam nanolithography system. For GaN PC slabs fabricated by dry etching, the light extraction was evaluated by measurement of the photoluminescence. The intensities of photoluminescence spectra were enhanced with the presence of GaN PC slabs as compared to that of the sample without PC. An exception to this occurred at 450 nm on the spectrum, for the PC with lattice constant of 230 nm, corresponding to the lattice condition of destructive diffraction at a half wavelength of light.

The triangular lattice PC structures were designed and fabricated on the top surface of a blue LED structure, as shown in Fig. 1. Epitaxial layers of blue LED structure were grown on sapphire substrates by using metal-organic chemical vapor deposition. After a 30-nm-thick buffer layer was grown at 520 °C, a 3-μm-thick *n*-type GaN layer was deposited at 1130 °C. Next we produced a multiple quantum well active region consisting of 2-nm-thick InGaN wells and 8-nm-thick GaN barriers emitting light at a blue wavelength of 450 nm. The multiple quantum well region consists of eight alternating layers of InGaN wells and GaN barriers, each grown at the same temperature of 790 °C. Then, a 250-nm-thick *p*-type GaN layer is grown and postannealed at 700 °C for 15 min.

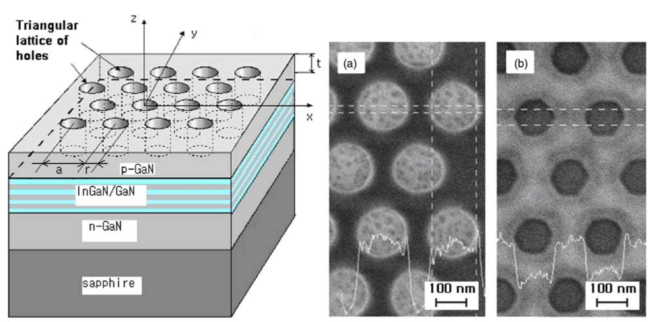

FIG. 1. (Color online) Schematic diagram of photonic crystal on the InGaN/GaN multi-quantum-well LED structure. The CD-SEM images for (a) the photoresist pattern obtained using electron beam nanolithography on the PMMA and (b) the GaN PC pattern after ICP etching process and the subsequent removal of residual PMMA.

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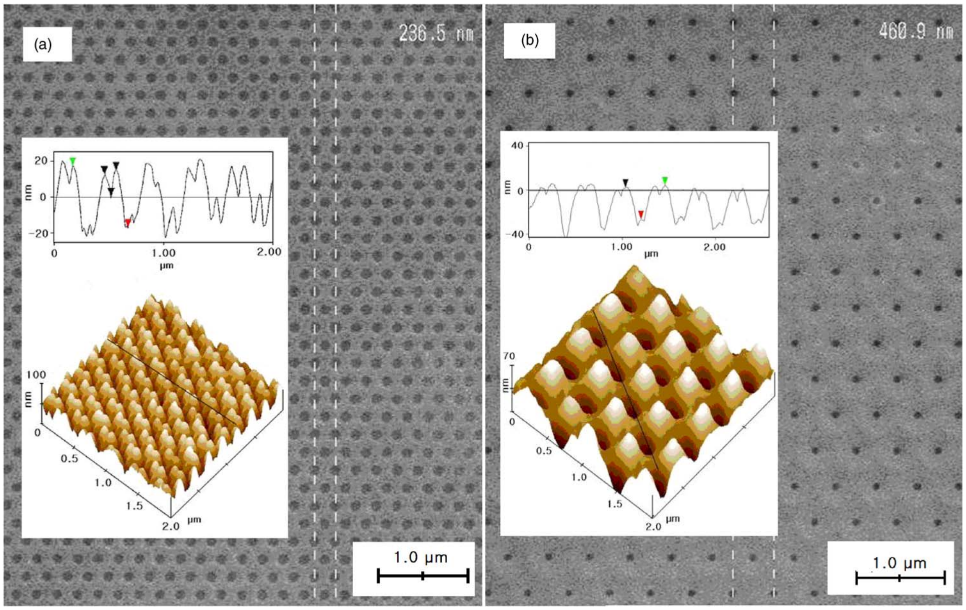

FIG. 2. (Color online) CD-SEM and three-dimensional AFM images for the samples of (a) PX230 and (b) PX460. From the sample of PX230, the images showed nanocolumns surrounding the holes for the lattice constant of 230 nm and the artificial hole diameter of 223 nm at surface and the height of 31 nm. For the sample of PX460, the hole diameter is about 218 nm at the top surface and the average depth of tapered holes is about 27 nm.

Considering the margin of the inductively coupled plasma (ICP) etching process, we initially designed samples in electron beam nanolithography: PX230 with a lattice constant of $a=230$ nm for the hole diameter of 150 nm and PX460 with a lattice constant of $a=460$ nm for the hole diameter of 150 nm. The GaN sample was coated with a 200-nm-thick PMMA photoresist and was processed to break out the polymerization of PMMA on the sites of the holes by electron beam nanolithography. The hole patterns of triangular lattices on the PMMA thin film were formed for a circular, 150 nm diameter with lattice constants of 230 and 460 nm on each $500 \times 500\ \mu m^2$ area in the same epitaxial wafer of blue LED. The patterned PMMA layer on the GaN surface was then developed in a solution of methylisobutyl ketone and isopropyl alcohol. The critical dimension–scanning electron microscopy (CD-SEM) images showed both the photoresist pattern, obtained using the electron beam nanolithography and the GaN PC patterns, after the ICP etching process and subsequent removal of residual PMMA.

The PMMA layer on the coated surface protects the GaN layer from etching by the plasma gas. The ICP dry etching process was performed for 5 s using the high density plasma consisting of a mixture of BCl_3 and Cl_2. The deep trench was formed with the air hole at the bottom having a diameter of 108 nm and with the stepped ring area having a diameter of 150 nm. The GaN photonic crystals were formed with a very rough and craterlike pattern at the top surface due to the ICP etching process. This indicates that the etching gas was isotropically dispersed at the surrounding area of the air holes, and the circular edges of PMMA film were etched out. The surface roughness and the depth of holes can be characterized by atomic force microscope (AFM) measurements, which can also be used to visualize the morphological formation of the spots and veins in the hexagonal array of air holes.

Figure 2(a) shows the CD-SEM and three-dimensional AFM images for the PX230 sample with a lattice constant $a=230$ nm. The CD-SEM image shows the hexagonal symmetry of nanoholes on a GaN surface. The holes were etched out to a diameter of 108 nm in the bottom areas as per a plane-view SEM image. The AFM image shows nanocolumnar structures surrounded by nanoholes. This means that the hole-surrounding area was disconnected due to the etching-beam scattering at the hole edges for the sample, which had a high occupation ratio of $r/a=0.485$. The artificial circular holes relatively surrounded by nanocolumns had diameters of 223 nm at the top surface. The average diameter and depth of the tapered air holes were 178 and 31 nm, respectively.

The nanocolumnar structures provide the light interaction media for the wave propagation. The dielectric constant shows the gradual change at the interfacial boundary region. The transition range of the refractive index at the interface is $\Delta r/a=0.05$ or 11.5 nm for $a=230$ nm. The refractive index of the PC can be reduced to the mean value between the dielectric material and the air, so that the pathway of light propagation can be smoothly established to enhance the light extraction onto surface. However, the geometric pathway of the light propagation can be blocked due to scattering from the PC because the Bragg diffraction condition forms a destructive interference to obstruct the light propagation.[10]

Figure 2(b) shows the CD-SEM and three-dimensional AFM images for the PX460 sample with lattice constant of $a=460$ nm. The CD-SEM image shows a hexagonal symmetry of holes and a dimple-shaped GaN surface around the holes. The holes were etched into a diameter of 76 nm at the bottom areas from the plane-view SEM image. The AFM image shows a nanohole array with triangular spikes surrounded by holes. The hole diameter is about 218 nm at the top surface and the average depth of tapered holes is about 27 nm. The PC structure has a hole occupation ratio of $r/a=0.237$. Therefore, after the ICP etching process, the triangular lattice PC was fabricated with an enlarged diameter of air holes and with a three-dimensional surface morphology as PC slabs. The optical properties of the fabricated PCs on the samples of PX230 and PX460 were characterized.

Room-temperature photoluminescence (PL) spectra from PCs are shown in Fig. 3. The PL spectra were measured with an incident of 40 mW He–Cd laser source activated at a 325 nm wavelength. For the bare sample (EPI) without 2D PC slab layer, the blue light emitted from the InGaN/GaN multiple quantum well structure is vertically incident to the p-type GaN surface layer, and it shows a very high PL intensity at 450 nm. Furthermore, the tiny PL peak at 364 nm

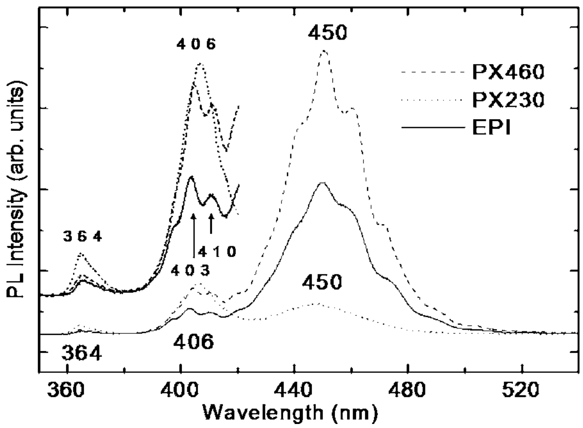

FIG. 3. Photoluminescence spectra from the InGaN/GaN multi-quantum-well LED structures. The PL from the bare sample (EPI) without PC showed the main peak at 450 nm and minor peaks at 364, 403, and 410 nm. The light extraction at 450 nm in the sample of PX230 is strongly suppressed due to the destructive Bragg diffraction and the PL intensity is smaller than that at 406 nm.

from the GaN band-to-band transition and the shoulder peaks at 403 and 410 nm from the Mg-doped *p*-type GaN layer have been observed.

For the sample of PX230, the PL intensity at 450 nm corresponding to the normal frequency $(a/\lambda=\omega a/2\pi c)$ of 0.511 was strongly suppressed, and the tiny PL bands of both 364 and 406 nm with normal frequencies of 0.631 and 0.566, respectively, showed enhanced light extraction. For the sample of PX460, the PL intensity at 450 nm with the normal frequency of 1.022 was strongly enhanced and even doubled. Furthermore, the 364, 403, and 410 nm PL bands with normal frequencies of 1.262, 1.141, and 1.122, respectively, showed enhanced intensities, though less than those in the sample of PX230. Therefore, for the same lattice array of photonic crystal, the light extraction is dependent on the wavelength of light emission, and the light extraction is also dependent on the lattice constant of the 2D thin PC slab under the same wavelength of light. This can be analyzed by wave optics.

Generally, in wave optics, the light incident to an array of similar apertures or nanoholes in a vertical plane can produce the Fraunhofer diffraction pattern. This pattern is given by the product of the diffraction pattern from a single nanohole and the interference pattern of an identically distributed array of point sources. This array of identical nanoholes provides a diffraction grating in wave optics, and the array theorem explains the interference from the nanograting. However, the theoretical estimation on the 2D PC only has the complete photonic band gap (PBG) for infinite thickness.[11] The triangular 2DPC slab with a finite thickness does not form the complete PBG for in-plane wave propagation. The rod and the hole 2D PC slabs provide the PBG for the transverse-magnetic (TM)-field-like and the transverse-electric (TE)-field-like polarization modes, respectively.[12,13] The complete PBG, for both TE and TM polarization modes, is possible for the three-dimensional PC.[14]

Our 2D PC slabs for LED devices are different from the 2D PC slabs for waveguides, where the photon propagates in the plane of nanohole array, but similar to the nanodiffraction gratings, where the photon is vertically incident to the plane. When the photon can propagate into the uniform dielectric media vertically to the plane of nanohole array, both components of electric field and magnetic field can be simultaneously placed in the plane of the array and can be scattered by nanoholes. The sample of PX230 having the normal frequency of 0.511 with the hole occupation ratio of 0.485 can confine the photon with wavelength of 450 nm, in the nanograting mode of destructive interference, from the photon scattering with PC. The sample of PX460 having the normal frequency of 1.022 with the hole occupation ratio of 0.237 can interact with the photon for the wavelength of 450 nm in the nanograting mode of constructive interference.

In summary, 2D photonic crystals were fabricated on a Mg-doped *p*-type GaN semiconducting thin film layer grown on eight alternating InGaN/GaN multi-quantum-well active layers by using metal-organic chemical vapor deposition. We experimentally showed the behavior of the light extraction of the 2D photonic crystal slab, which depended on the lattice constant and the emission wavelength of the light. The photonic crystal slab affects vertically incident light for lattice constant of about half a wavelength, which corresponds to destructive interference in the diffraction grating. As a concluding remark, the two dimensional nanohole array provides nanodiffraction for vertically incident light as a quantum scattering of light in photonic crystal structure.

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