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Abstract

Instead of using conventional electron lithography, a two-dimensional photonic crystal consisting of a hexagonal array of triangular air-holes was created on the surface of a GaN LED substrate using microsphere lithography. The microspheres self-assemble into a single-layered hexagonal-close-packed array acting as an etch mask. A significant enhancement in photoluminescence intensity was recorded from the PhC LED structure. A twofold increase in electroluminescence was observed from the PhC LED compared to an as-grown LED with identical geometry. Besides geometrical factors due to surface roughening, the dispersive nature of PhCs and diffractive properties of the PhC as a grating contribute to the enhancement of light extraction from the LED.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Direct wide-bandgap gallium nitride (GaN) has become a prominent material in the optoelectronics market, with numerous evolutionary products that have been commercialized in recent years, including the blue laser diode [1] for high-density optical storage and the white light-emitting diodes (LEDs) for general lighting purposes. While earlier research efforts have been focused on improving material epitaxy, there has been growing concern over the limited fraction of light that can be extracted as a result of the large refractive index contrast. Improving light extraction efficiency of LEDs has thus become a topic of considerable interest. Incorporation of micro-and nanoscale features, in various geometries of discs, rings and hexagons, into GaN LEDs have previously been reported to promote light extraction by various extents [2, 3]. On the other hand, random surface roughening of a GaN surface via a self-assembled Ni nanomask has been demonstrated to improve light output by 1.55 times whilst reducing p-contact resistance [4]. However, such methods merely rely on geometric effects to facilitate light extraction. Photonic crystals (PhC) [5–12] constitute another promising candidate towards improving light extraction of LEDs. A PhC structure consists of a periodic arrangement of two media with different dielectric constants of the order of the wavelength of light, forming two interpenetrative domains. A range of device structures comprising PhCs have been proposed and demonstrated to boost their optical performances via the various properties of PhCs.

There are two major schemes in which PhCs can be exploited for LED applications, the first being the formation of photonic bandgaps to prevent emission into guided modes [13]; the other involves the dispersive nature of PhC structures to extract guided light [14, 15]. In this paper, we report on an implementation of the second scheme.

Currently, severe challenges exist in fabricating PhCs effectively. Conventional nanopatterning has relied heavily on traditional techniques such as electron-beam lithography for optoelectronic devices. The drawbacks of e-beam writing are high cost and low throughput, making it unsuitable for mass production. On the other hand, the resolution of DUV lithography is limited by diffraction effects, despite its higher throughput. Nano-imprinting is a promising technique, albeit with a hefty price tag. This situation encourages the development of alternative self-assembly methods for nanostructure patterning suitable for mass production. We present a simple, low cost and efficient method of fabricating a two-dimensional PhC on the surface of GaN LEDs using the technique of microsphere lithography. Details of the fabrication and characterization of a photonic crystal LED based on this controllable and economical method are reported.

2. Experimental details

Spontaneous assembly of microspheres involves the spreading of the micrometer-scale spherical particles uniformly across
a substrate. This is achieved by dispensing microspheres suspended in deionized water onto a sample, followed by a spin-coating process. Using appropriate process parameters, the microspheres self-assemble into a single-layered hexagonal-close-packed array. In this work, silica (SiO$_2$) microspheres, having mean diameters of 500 nm with a uniformity of better than 1%, were used to form the lithographic mask, which subsequently act as an etch mask for pattern transfer onto the GaN LED substrate, whose structure consists of In$_{x}$Ga$_{1-x}$N/GaN multi-quantum wells (MQWs) grown by metal–organic chemical vapor deposition (MOCVD) with center emission wavelength of 470 nm. Details of the assembly process can be found in [16]. A plan view of a close-packed microsphere monolayer array imaged by field-emission scanning electron microscopy (FE-SEM) is shown in figure 1.

The microsphere-coated samples were dry etched by inductively coupled plasma (ICP) etching using the gas chemistry of Cl$_2$ and BCl$_3$, producing an etch rate of 490 nm min$^{-1}$ and an etch selectivity of 4:1 between GaN and the SiO$_2$ microspheres. The void regions between the microspheres allow plasma penetration, creating air-holes on the semiconductor surface. The silica particles were subsequently removed by sonification. Triangular air-holes of 180 nm on a side with a spacing of 290 nm were formed on the top region, corresponding to a periodic two-dimensional semiconductor–air grating on the planar surface. By adjusting the etch duration, triangular air-holes of different depths can be obtained. PhCs with different hole dimensions and spacings can be obtained with microspheres of different diameters.

Being a systematic and reproducible technique based on spin-coating, large areas of microspheres consisting of monolayers can be formed. In our experiments, samples of 10 mm x 10 mm dimensions are routinely used; monolayer coverage usually exceeds 70%, except for an edge bead at the periphery of the sample. Alternative coating methods are being explored to further improve coverage, including vertical deposition techniques, with the target of reducing defect density and increasing monolayer coverage [17].

Micro-photoluminescence (µ-PL) spectra were collected to evaluate the optical properties of the PhC structures, using a Spectra-Physics diode-pumped solid state (DPSS) ultraviolet (UV) laser at 349 nm as an excitation source. The beam was focused to a spot of about 10 µm with a triplet lens, and the PL signal was collected and coupled to a spectrometer via an optical fiber. An electroluminescent LED was subsequently fabricated using the PhC template.

3. Results and discussions

3.1. Enhancement of PL intensity in PhC structures

To evaluate the effects of PhC incorporation, different etch depths were investigated. The first PhC template is dry etched for 30 s, corresponding to an etch depth of ~250 nm in the unmasked open regions, as shown in figure 2(a). Based on previous cross-sectional measurements, the depths of the air-holes were approximately 1/2 of this value, that is, ~125 nm. A second sample was etched for 45 s, with an estimated air-hole penetration of ~190 nm, as shown in figure 2(b). The µ-PL spectra from these two samples were collected, together with that from the as-grown sample, which are plotted collectively in the graph of figure 3. As evident from the plot, the PL intensity was enhanced with the PhC grating, and scales with the depth of the air-holes. Clearly, the PhC array is promoting light extraction from the LED device. To further study the mechanism involved, complete LED structures were fabricated.

3.2. Fabrication of PhC LEDs

Light-emitting diodes were subsequently fabricated using the PhC template with air-holes of ~125 nm depth, following the fabrication sequences depicted in figures 4(a)–(d). A semi-transparent current spreading layer consisting of an Ni/Au bi-layer (10 nm/10 nm) was deposited by e-beam
evaporation onto the template via a photoresist lithographic mask containing multiple developed regions of 150 μm by 150 μm, thus defining the light emissive areas, which are well aligned to the PhC arrays. The metal layer was lifted off and annealed at 550°C for 15 min in ambience to form ohmic contacts, as depicted in figure 4(b). The sample was then dry etched by ICP to remove the MQWs and to expose the n-region, as shown in figure 4(c). This is followed by depositing the Al/Ti (40 nm/300 nm) contact pads by electron-beam deposition with subsequent annealing at 550°C for 5 min in nitrogen. The cross-sectional and three-dimensional schematic diagrams of the PhC LED structure are shown in figures 4(d) and 5(a), respectively.

3.3. Enhancement of EL intensity in PhC LEDs

The fabricated PhC LED with light emission area of 150 nm × 150 nm is shown in the FE-SEM image in figure 5(b). A device of identical geometry, except without the PhC array, was fabricated alongside for comparison. Electroluminescence (EL) of the devices was demonstrated at room temperature in probe measurement geometry and the corresponding optical spectra were collected via an optical fiber placed close to the devices. Figure 6 shows the EL spectra of the PhC LED and as-grown LED biased at the injection current of 1.5 mA. The devices were operated at relatively lower currents for their smaller dimensions. From the spectra, the EL intensity is enhanced.
~1.8 times compared to the as-grown LED. A minor blueshift was observed, together with the disappearance of interference fringes in the spectrum of the PhC LED. The plot of EL intensity versus injection current is plotted in figure 7, whereby it is observed that the slope of the curve for the PhC LED is higher than that of the as-grown LED, indicating its higher efficiency. Plan view microphotographs of the devices driven at 0.5 mA are illustrated in figure 8; the PhC LED distinctively delivers higher optical output than the as-grown LED.

The origins of the optical enhancement through the PhC can be explained from various points of views. The presence of air-holes converts the otherwise smooth surface into a roughened surface, which promotes light extraction via geometrical effects. The large refractive index of GaN of $n = 2.5$ at 470 nm [18] restricts light extraction to an emission cone of about 30°. In other words, as much as 80% of photons generated by the MQWs remain trapped within the device structure. The introduction of air-holes onto the surface offers a large surface area for photon out-coupling via a wider range of emission angles. On top of geometrical considerations, the optical properties of PhCs play a major role in enhancing light emission. There are two major properties associated with a PhC array, namely the photonic bandgap and dispersion effects. In this implementation the photonic bandgap is not relevant; the presence of a bandgap would impede light transmission across the PhC. Instead, its dispersive properties give rise to a modulation of the refractive index, similar to the effects of having an index matching layer deposited on the top of the LED, thus expanding the light emission cone.

Since the dimension of the PhC structure is of the order of the wavelength of light, light diffraction will occur as it transmits through the PhC, causing light waves to bend and emit in a different direction. Such Bragg scattering allows photons to escape from the GaN film and also increases the directionality of the LED radiation pattern. In the as-grown LED, most of the energy is emitted into waveguided modes internal to the semiconductor rather than radiation modes, due to reflections between the GaN/sapphire and GaN/air interfaces. Light generated inside the LED bounces to and fro, setting up standing waves, as evidenced through the interference fringe patterns in the spectra of figure 6 (for the as-grown LED). Most of these photons are re-absorbed before they can escape from the semiconductor. The introduction of PhCs can improve light extraction by diffracting waveguided modes out of the semiconductor, with the EL spectrum shown in figure 6 (for the PhC LED) providing strong evidence through the disappearance of modal features.

One of the concerns pertaining to self-assembly techniques is the formation of defects in the array (including point and line defects). Defect formation in the microsphere assembly procedure is analogous to the crystal growth process. From figure 8, it is observed that light emission through the PhC grating is not entirely uniform; it is attributed to the presence of defect clusters. Various point defects are revealed in the SEM image in figure 9, which shows a close-up image of the emission area of the PhC LED with defects. Coincidentally, the peripheries of such defect clusters appear brighter than other regions in the SEM image, which indicate an accumulation of electrons at the edges of defects. This phenomenon offers a clue on the effects
of defect generation in the PhC array. The existence of such defects breaks the periodicity of the photonic crystal structure, hence affecting its overall light-guiding capability. When photons encounter these defects, they will leak out from the photonic crystal structure into these localized modes, reducing its light-guide capacity. Moreover, the locations of defects are exposed to the plasma during dry etching and are subjected to plasma damage [19]. Since the formation of ohmic contact to a plasma-damaged p-type GaN surface results in high contact resistivities, the light output from these regions are expected to be lower, resulting in the inhomogeneous light distribution from the emission area.

4. Conclusions

In summary, we have demonstrated the fabrication of a GaN LED with a PhC incorporated on the surface. The PhC was patterned with microsphere lithography, a technique involving the self-assembly of spherical particles into hexagonal-close-packed arrays. Upon dry etching, air-holes are formed in the void regions between the microspheres. PL measurements reveal a threefold enhancement of the light emission intensity due to increase in light extraction efficiency. LEDs were subsequently fabricated using the photonic crystal template and the EL measurements exhibited an improvement of light emission by up to twofold. This significant performance boost can be attributed to various factors, including geometrical scattering due to a roughened surface, light dispersion of the photonic crystal and light diffraction through the PhC grating.

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References

[7] Ichikawa H and Baba T 2004 Appl. Phys. Lett. 84 457