Close-packed hemiellipsoid arrays: A photonic band gap structure patterned by nanosphere lithography

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A self-assembled hexagonal close-packed hemiellipsoidal photonic crystal structure was fabricated on GaN material. An ordered monolayer silica nanosphere coating served as a hard mask in an inductively coupled plasma etching process. The shape of the arrayed hemiellipsoids can be controlled by adjusting the etch selectivities and durations according to the fabrication model. The existence of a photonic band gap is established through planar transmissivity measurement whereby a transmission dip centered at 440 nm was identified. A threefold enhancement in light extraction was achieved, as determined from the measured angular photoluminescence emission pattern.

The proposed PBG structure was fabricated through a dual-step process, the process flow of which is illustrated in the schematic diagram of Fig. 1. A LED wafer, containing InGaN/GaN MQWs with emission centered at 440 nm, was initially coated with plain 192-nm-diameter SiO₂ nanospheres with coefficient of variation (CV) ~4% (acquired from Corpuclus Inc.) by vertical deposition method. The nanosphere solution, together with alcohol and sodium dodecyl sulfate, which accelerate the resulting solution’s evaporation rate and reduce viscosity, respectively, were mixed in a vial. The wafer was immersed into the solution by laying it upon the sidewall of the vial, which was then placed into an oven maintained at a constant temperature of 40 °C with a stable supply of air stream for 3 hrs. The solution eventually evaporated, leaving the nanospheres self-assembled into a monolayer of HCP pattern on the surface of the wafer.

Subsequently, the coated sample was subjected to ICP dry etching for pattern transfer, during which the monolayer nanosphere coating served as a hard mask. A monolayer coating of nanosphere is thus essential for pattern transfer. Inability of our proposed structure in enhancing light extraction efficiency are demonstrated.

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In this paper, we propose a self-assembled PhC structure based on a HCP hemiellipsoid (HE) array. The geometry of the HEs can be adjusted by controlling the process conditions (namely etch selectivity and duration), thereby tuning the PBG properties. The existence of a PBG and the feasibility of our proposed structure in enhancing light extraction efficiency are demonstrated.

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Fig. 2 was obtained after a total etch duration of 290 s, as shown in
structure etched for a duration of $t$ can be deduced according
to the equations and diagram in Fig. 2(a), with $E_{\text{GaN}}$ and $E_{\text{SiO$_2$}}$ being the etch rates of GaN and SiO$_2$, respectively.

The HE structure was progressively shaped during the
dry etch process. The field-emission scanning electron micro-
scope image in Fig. 2(b) illustrates nanopillars with slightly inclined sidewalls formed after etching for 120 s, at
gas flow rates of 12 SCCM (SCCM denotes cubic centimeter
per minute at STP) for Cl$_2$ and 9 SCCM for CHF$_3$, respec-
tively. Based in this recipe, the etch selectivity between GaN and SiO$_2$ was approximately 4:3. The final required structure
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Fig. 2(c). The nanosphere residue was removed by 10 min of
sonication in acetone. The height of the resulting structure is
roughly 250 nm, leaving a gap of about 50 nm between the
bottom of the HE structure, and also from the triangular area
which represents the near field extracted light energy flux.
The result shows that most of the energy is extracted at the
bottom of the HE structure, and also from the triangular area
among the HE. Such periodic spatial emission pattern sug-
gests the diffractive nature of light emission from the PhC
structure. Also, the results show 88.6% of guided modes in
the $p$-GaN layer are coupled out.

A finite-difference time-domain (FDTD) simulation was
also carried out to predict the energy flux distribution across
the PBG structure. The simulation, coded in MATLAB, was set
to have a dipole excitation source emitting at 440 nm, with
symmetric boundary in all directions except in the upward
direction, where the perfectly matched layer was imple-
mented. Figure 3(d) is the accumulated energy flux distribu-
tion sampled in the air medium just above the GaN structure,
which represents the near field extracted light energy flux.
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To verify the existence of a PBG, a planar transmission
measurement was conducted. Light emitted from a broad-
bond tungsten-halogen lamp was collected by a fiber. After
being collimated by two plano-convex lenses, the beam was
focused by a 20× magnification UV objective. With slight
downward inclination, the beam was directed onto the
sample exciting guiding modes within the wafer, resulting in
three output beam spots, corresponding to the three guiding
modes along the $p$-$\text{GaN}$, $n$-$\text{GaN}$, and sapphire layers. The
output light beam corresponding to the guiding mode be-
tween the $p$-$\text{GaN}$ layer and MQW, where the PBG structure
is situated, was then collected and coupled by another optical
fiber to a spectrometer. Figure 4(a) shows the measured
transmission spectrum. A pronounced transmission dip cen-
tered around 440 nm with a full width at half maximum
of 55 nm was observed, correlating well with the simulated
results, testifying to the existence of a PBG. The slight de-
viation may be attributed to defects, which include line defects, point defects, and shape irregularities that exist within the self-assembled PhC, and possibly due to size deviation of nanospheres from the expected value, although these defects may actually contribute to better coupling efficiency due to the introduction of defect modes. Besides, fringes in the spectrum are attributed to the Fabry–Perot (FP) modes within the GaN layer in the reference spectrum taken from an as-grown sample. The maximum transmission was below 100%, probably due to light scattering caused by the PhC structure.

A PBG structure integrated into a LED promotes light extraction efficiency in two major ways. The presence of a PBG in the direction of the plane suppresses laterally propagating guiding modes, channeling them into the vertical direction across the GaN-air interface. It also gives rise to a surface texturing effect. Angle-resolved photoluminescence (PL) measurements were carried out to evaluate the possible effects of a PBG structure on the emission pattern. The beam from a third harmonic neodymium: yttrium lithium fluoride (Nd:YLF) diode-pumped solid state laser from Spectra-Physics at wavelength of 349 nm was focused onto the sample at an oblique angle to excite the MQWs beneath. The high absorption UV photons in GaN prohibits light guiding, restricting the excitation to a tiny spot. The emitted light was then collected by a fiber at a constant separation of 20 mm directly on top of the excited spot, at angles varying from 0° to 90° at a 1° interval. The measured angular PL emission pattern and angle-resolved PL spectrum are presented in Fig. 4.

According to Fig. 4(d), a significant overall increase in light extraction efficiency was observed, with an enhancement factor of more than 3, computed by comparing the integrated PL intensities of the emission patterns of the patterned and as-grown samples. The light intensity for the as-grown sample starts to drop as it approaches the critical angle of GaN and air (of 23°), while the sample with the PBG structure maintains considerable light intensity up to 60°. This is attributed to surface texturing effect brought about by the PhC geometry, since the array of HEs acts as an effective medium for light to escape, producing a batwing emission pattern. This is consistent with the FDTD simulation of Fig. 3(d) that light emission will be highly diffracted. Also observed in Fig. 4(b), the angle-resolved PL spectrum of the as-grown sample contains fringes in the diagonal direction. These fringes correspond to shifts in the FP modes due to optical oscillations in the GaN layer. Similar fringes in Fig. 4(c) are significantly suppressed as the FP modes are annihilated due to the surface texturing effect introduced by the PBG structure. On top of this, light extraction is further enhanced via the PBG. Vertical fringes observed between 0° and 20° in Fig. 4(c) can be attributed to the diffraction due to PhC. No emission lines are exhibited in Fig. 4(c), since the PBG covers a wavelength range of 420 to 480 nm, as demonstrated in Figs. 3(a) and 4(a).

Secondary effects observed from the PL spectrum are also associated with the surface nanostructuring. First, the primary emission peak exhibits a spectral blueshift of 10 nm, attributed to higher effective optical pumping density caused by the enhanced light coupling efficiency after implementing the PBG structure, despite the optical pumping densities at the source being unchanged for both measurements. Additionally, an extra emission peak is observed at about 395 nm, due to exposure of the Mg-doped AlGaN current blocking layer embedded between the MQW and p-GaN layer after etching.

Even with a threefold enhancement in light extraction achieved, there is further room for improvement by optimizing light coupling between the MQW and the PBG structure, which has not been addressed in this paper. The etch depth was intentionally chosen to minimize damage to MQWs by plasma exposure but a deeper penetration extending beyond MQW region may form a nanoresonant cavity which can improve the Purcell factor as a result of a greatly reduced effective volume, and a high cavity quality factor due to PBG. The HE geometry presented in this work distinguishes from other common flat-topped PhC structures in terms of light extraction, bearing resemblance to the highly effective hemispherical encapsulation albeit on a nanoscale.

In summary, enhanced light extraction with a self-assembled arrayed HEs PBG structure has been demonstrated. The shape of the self-assembled array can readily be controlled through adjusting the etching parameters, and thus resulting in tunability of the PBG position. Through optical transmission and angle-resolved PL experiments, the predicted TE-mode PBG is verified, which was proven to play a vital role in promoting light extraction.