Light extraction efficiency enhancement of InGaN quantum wells light-emitting diodes with polydimethylsiloxane concave microstructures

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Abstract: Improvement of light extraction efficiency of InGaN light emitting diodes (LEDs) using polydimethylsiloxane (PDMS) concave microstructures arrays was demonstrated. The size effect of the concave microstructures on the light extraction efficiency of III-Nitride LEDs was studied. Depending on the size of the concave microstructures, ray tracing simulations show that the use of PDMS concave microstructures arrays can lead to increase in light extraction efficiency of InGaN LEDs by 1.5 to 2.0 times. Experiments utilizing 2.0 micron thick PDMS with 1.0 micron diameter of the PDMS concave microstructures arrays demonstrated 1.70 times improvement in light extraction efficiency, which is consistent with improvement of 1.77 times predicted from simulation. The enhancement in light extraction efficiency is attributed to increase in effective photon escape cone due to PDMS concave microstructures arrays.

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References and links

- 1. Need Project, Intermediate Energy Infobook, (US Department of Energy, pp. 51, 2008).
- 2. E. F. Schubert, Light Emitting Diodes, (Cambridge University Press, pp. 145, 2006).
- 3 S. Nakamura, and S. Pearton, S., and G. Fasol, The Blue Laser Diode, (Springer-Verlag Berlin Heidelberg, pp. 7, 2000).
- R. A. Arif, Y. K. Ee, and N. Tansu, "Polarization Engineering via Staggered InGaN Quantum Wells for 4. Radiative Efficiency Enhancement of Light Emitting Diodes," Appl. Phys. Lett. **91**(9), 091110 (2007). R. A. Arif, H. Zhao, Y. K. Ee, and N. Tansu, "Spontaneous Emission and Characteristics of Staggered InGaN
- 5 Quantum Wells Light Emitting Diodes," IEEE J. Quantum Electron. 44(6), 573-580 (2008).
- H. Zhao, R. A. Arif, and N. Tansu, "Design Analysis of Staggered InGaN Quantum Wells Light-Emitting Diodes at 500-540 nm," IEEE J. Sel. Top. Quantum Electron. 15, 1104-1114 (2009).
- H. Zhao, R. A. Arif, and N. Tansu, "Self Consistent Gain Analysis of Type-II 'W' InGaN-GaNAs Quantum Well 7. Lasers," J. Appl. Phys. **104**(4), 043104 (2008). H. Zhao, R. A. Arif, Y. K. Ee, and N. Tansu, "Self-Consistent Analysis of Strain-Compensated InGaN-AlGaN
- 8 Quantum Wells for Lasers and Light Emitting Diodes," IEEE J. Quantum Electron. 45(1), 66-78 (2009).
- C. Huh, K. S. Lee, E. J. Kang, and S. J. Park, "Improved light-output and electrical performance of InGaN-based light-emitting diode by microroughening of the p-GaN surface," J. Appl. Phys. 93(11), 9383-9385 (2003).
- 10. T. Fujii, Y. Gao, R. Sharma, E. L. Hu, S. P. DenBaars, and S. Nakamura, "Increase in the extraction efficiency of GaN-based light-emitting diodes via surface roughening," Appl. Phys. Lett. 84(6), 855-857 (2004).
- 11. C. F. Lin, Z. J. Yang, J. H. Zheng, and J. J. Dai, "Enhanced light output in nitride-based light-emitting diodes by roughening the mesa sidewall," IEEE Photon. Technol. Lett. 17(10), 2038-2040 (2005).
- 12. S. J. Lee, J. Lee, S. Kim, and H. Jeon, "Fabrication of reflective GaN mesa sidewalls for the application to high extraction efficiency LEDs," Phys. Stat. Solidi (c) **4**, 2625-2628 (2007).
- T. Kim, A. J. Danner, and K. D. Choquette, "Enhancement in external quantum efficiency of blue light-emitting diode by photonic crystal surface grating," Electron. Lett. **41**(20), 1138–1139 (2005). 14. J. J. Wierer, M. R. Krames, J. E. Epler, N. F. Gardner, J. R. Wendt, M. M. Sigalas, S. R. J. Brueck, D. Li, and M.
- Shagam, "III-nitride LEDs with photonic crystal structures", in Proc. SPIE, 5739, 102-107 (2005).

- K. McGroddy, A. David, E. Matioli, M. Iza, S. Nakamura, S. DenBaars, J. S. Speck, C. Weisbuch, and E. L. Hu, "Directional emission control and increased light extraction in GaN photonic crystal light emitting diodes," Appl. Phys. Lett. 93(10), 103502 (2008).
- H. W. Choi, C. Liu, E. Gu, G. McConnell, J. M. Girkin, I. M. Watson, and M. D. Dawson, "GaN micro-lightemitting diode arrays with monolithically integrated sapphire microlenses," Appl. Phys. Lett. 84(13), 2253–2255 (2004).
- J. Q. Xi, H. Luo, A. J. Pasquale, J. K. Kim, and E. F. Schubert, "Enhanced light extraction in GaInN lightemitting diode with pyramid reflector," IEEE Photon. Technol. Lett. 18(22), 2347–2349 (2006).
- J. K. Kim, M. F. Schubert, J. Q. Xi, F. W. Mont, and E. F. Schubert, "Enhancement of light extraction in GaInN light-emitting diodes with graded-index indium tin oxide layer", in *Proc. of the Conference on Lasers and Electro-Optics* (Baltimore, MD, 2007) Paper No. CTuI1.
- J. Q. Xi, M. F. Schubert, J. K. Kim, E. F. Schubert, M. Chen, S. Y. Lin, W. Liu, and J. A. Smart, "Optical thinfilm materials with low refractive index for broadband elimination of Fresnel reflection," Nat. Photonics 1, 176– 179 (2007).
- A. J. Fischer, F. W. Mont, J. K. Kim, E. F. Schubert, D. D. Koleske, and M. H. Crawford, "Enhanced Light-Extraction from InGaN Quantum Wells Using Refractive-Index-Matched TiO₂", in *Proc. of the Conference on Lasers and Electro-Optics* (Baltimore, MD, 2007) Paper No. CTuI2.
- Y. K. Ee, P. Kumnorkaew, R. A. Arif, J. F. Gilchrist, and N. Tansu, "Enhancement of light extraction efficiency of InGaN quantum wells light emitting diodes using SiO₂/polystyrene microlens arrays," Appl. Phys. Lett. 91(22), 221107 (2007).
- P. Kumnorkaew, Y. K. Ee, N. Tansu, and J. F. Gilchrist, "Investigation of the deposition of microsphere monolayers for fabrication of microlens arrays," Langmuir 24(21), 12150–12157 (2008).
- Y. K. Ee, P. Kumnorkaew, R. A. Arif, H. Tong, H. Zhao, J. F. Gilchrist, and N. Tansu, "Optimization of Light Extraction Efficiency of III-Nitride Light Emitting Diodes with Self-Assembled Colloidal-based Microlenses," IEEE J. Sel. Top. Quantum Electron. 15, 1218–1225 (2009).
- E. H. Park, J. Jang, S. Gupta, I. Ferguson, C. H. Kim, S. K. Jeon, and J. S. Park, "Air-voids embedded high efficiency InGaN-light emitting diode," Appl. Phys. Lett. 93(19), 191103 (2008).
- R. Horvath, L. R. Lindvold, and N. B. Larsen, "Fabrication of all-polymer freestanding waveguides," J. Micromech. Microeng. 13(3), 419–424 (2003).
- B. G. Prevo, and O. D. Velev, "Controlled, rapid deposition of structured coatings from micro- and nanoparticle suspensions," Langmuir 20(6), 2099–2107 (2004).
- 27. J. E. Mark, Handbook of Polymers, (Oxford University Press, pp. 424, 1999).
- J. Zou, D. Kotchetkov, A. A. Balandin, D. I. Florescu, and F. H. Pollak, "Thermal conductivity of GaN films: Effects of impurities and dislocations," J. Appl. Phys. 92(5), 2534–2539 (2002).
- N. Tansu, and L. J. Mawst, "Current Injection Efficiency of 1300-nm InGaAsN Quantum-Well Lasers," J. Appl. Phys. 97(5), 054502 (2005).

1. Introduction

The rising energy cost has pushed for technological advances for high energy-efficiency technology. The United States spends more than \$37 billion annually on energy for lighting alone [1]. One of the long term strategies adopted to reduce the energy consumption is solid state lighting through the use of light emitting diodes (LEDs). The LED technology represents a disruptive technology that has the potential to displace its less energy efficient lighting devices such as incandescent and fluorescent lamps. High efficiency InGaN quantum wells (QW) LEDs [2–4] in particular have already been used in applications such as full color displays, traffic signals, backlighting for liquid crystal displays as well as solid state lighting.

The external quantum efficiency (η_{EOE}) of LEDs can be expressed as the product of

current injection efficiency (η_{inj}), radiative efficiency (η_{rad}), and light extraction efficiency

 $(\eta_{extraction})$). The light extraction efficiency is defined as the fraction of photons generated in

the active region of semiconductor that escapes into free space. In addition to challenges in achieving high radiative efficiency and current injection efficiency in InGaN QW LEDs [4–8], light extraction efficiency optimization in nitride-based LEDs is important for achieving high-efficiency devices. Unfortunately, the refractive index difference of GaN (n = 2.5) is relatively large compared to air (n = 1). The large refractive indices mismatch results in a narrow escape cone of only 23.5° with escape probability of only 4% from the top surface of the LED devices, with the rest of the optical power trapped within the semiconductor.

To overcome the light extraction limitation in nitride LED, several novel approaches have been pursued [9–20]. One approach is to roughen the top surface of the LEDs, which will lead to enhanced scattering of light generated from the active region at the roughened GaN / air

interface. The surface roughening technique could be accomplished by using wet etching [9] or photochemical etching [10], however the roughness obtained using these methods were not uniform thus leading to variation in the light extraction efficiency improvement of the LEDs across the wafer sample. Besides roughening the top surface emission area of the LEDs, other research groups have tried to roughen the mesa sidewalls of the LEDs using photochemical etching [11]. Similarly, the surface of the mesa sidewalls was non-uniform. Another approach to increase the light extraction efficiency of the LEDs was to use an oblique mesa sidewall [12], which can be achieved by using reflowed photoresist and adjusting the flow of CF_4 gas during the dry etch process. The improvement of the light extraction efficiency in LED employing oblique mesa sidewall [12] was isolated to the areas in the sidewall regions only. Photonic crystal structures can also be utilized to enhance the light extraction efficiency of LEDs [13–15], but this approach requires the use of electron beam or complex laser holographic lithography due to the small dimensions of the photonic crystal structures. Other approaches to improve light extraction efficiency in III-Nitride LEDs also include sapphire microlens approach [16], nanopyramid approach [17], and grading refractive indices between GaN / air interface with planar materials approach [18–20]. Recently, we demonstrated a novel approach to significantly enhance the light extraction efficiency of III-Nitride LEDs by fabricating SiO₂ / polystyrene (PS) microlens arrays [21–23].

Here, we present a novel technique to increase the light extraction efficiency of III-Nitride LEDs using polydimethylsiloxane (PDMS) concave microstructures arrays. In our previous work on SiO₂ / PS microlens arrays [21–23], we formed convex lens-like microstructures on top emission area of the LEDs by semi-burying SiO₂ microspheres in planar polystyrene by using rapid convective deposition method. In this work, specifically we employed SiO₂ microsphere arrays as a template to form concave microstructures arrays in the PDMS layer on top of the LEDs (Fig. 1) by using imprinting method. The use of imprinting method leads to a low cost and scalable approach to enhance the light extraction efficiency of the LEDs. The use of concave microstructures arrays, instead of convex microstructures, on top of LEDs also leads to improved self-focusing effect of the extracted photons. The size effect of the PDMS concave microstructures arrays on the light extraction efficiency of InGaN QWs LEDs has been studied. The experimental and simulation results will be compared with those of planar LEDs.



Fig. 1. Schematic of InGaN QWs LEDs structure utilizing PDMS concave microstructures arrays, on the top emitting InGaN LED.

2. Computation of light extraction efficiency via Monte Carlo ray tracing

To study the effect of PDMS concave microstructures arrays on the light extraction of InGaN QWs LED, simulation studies were conducted using Monte Carlo ray tracings. Monte Carlo technique was used to calculate the light extraction efficiency by treating the entire LED device structure as a physical optics problem, and tracking the rays emanating from the quantum wells active region of the device. Note that wave-optics based modeling will provide improved accuracy in the simulation of the structure. However, Monte Carlo ray tracing is sufficient for simulating our current device structure [24], due to the micron-sized features

employed in the our devices. The spontaneous emission of the InGaN QWs were computed and used as the light source in the ray tracing simulation. The interaction of the ray traced photons and carrier transport is carried out in three-dimensions self-consistently. Photon recycling recombination process was also taken into account in the simulation. The quantum wells active domain comprises of 74,000 vertices (20 rays in each vertex) with 1.48 million light rays used in the simulations to minimize statistical error to less than 0.1%. The schematic of the simulation mesh structure of the LED device utilizing the PDMS concave microstructures arrays is shown in Fig. 2(a).



Fig. 2. (a) Schematic of InGaN QWs LEDs simulation mesh structure utilizing PDMS concave microstructures arrays on the top emission surface, and (b) flow chart of Monte Carlo ray tracing simulation to calculate the light extraction efficiency of the LED.

The LED structure in the simulation comprises of a 100µm x 100µm mesa size LED device structure grown on 2.5µm n-GaN virtual template on 400µm thick sapphire substrate. The active region consists of 4 periods of 2.5nm InGaN/GaN QWs, capped with 0.1µm pdoped GaN used as the top contact layer. Finally, a layer of PDMS concave microstructures was designed on top of the emission region. The flow chart of the Monte Carlo simulation is shown in Fig. 2(b). First the physical LED structure was defined, together with the input material parameters such as refractive index. The refractive index of PDMS (Sylgard 184) used in the simulation was 1.43 [25]. Additional LED ray tracing parameters such as number of rays per vertex, minimum intensity of ray before raytracing terminates were also defined. Ray tracing of the photons starts with the spontaneous emission from the active region, which comprises of a 4-period InGaN QWs. Tracing of the trapped rays were terminated after a preset minimum intensity has been reached. Light extraction efficiency was then obtained from the analysis of the outgoing rays from the LED structure. To provide useful comparison in our studies, the ratio of the light extraction efficiency of the III-Nitride LED employing PDMS concave microstructure arrays with that of planar LED is compared for various microstructure diameter (d_{concave}) and PDMS layer thicknesses with various emission wavelengths.

To investigate the size effect of PDMS concave microstructures arrays on the light extraction efficiency of III-nitride LEDs, ray tracing simulation was done for LEDs with light emission area coated with PDMS concave microstructures arrays. Figures 3(a) and 3(b) show the ratios of the light extraction efficiencies of the LEDs with concave microstructures arrays compared to the planar LED, with PDMS layer thicknesses of 3.0µm and 2.0µm, respectively.

The ratio of light extraction efficiency improvements were investigated for LEDs emitting at 420nm, 480nm and 525nm with diameter of the PDMS concave microstructures arrays ($d_{concave}$) ranging from 0.3µm to 2µm. For the case of 3.0µm thick PDMS layer, the use of these concave microstructures arrays leads to increase in the light extraction efficiencies by 1.4 times up to 1.9 times for $d_{concave}$ ranging from 0.3µm up to 2µm as shown in Fig. 3(a). As the size of the concave microstructures increases ($d_{concave} > 1.5 \mu$ m), the improvement in light extraction efficiency for the LED employing PDMS concave microstructures arrays reduces and approaches to that of planar LEDs. The light extraction efficiency enhancement using PDMS concave microstructures arrays is relatively independent of the three different emission wavelengths investigated here. For LEDs with similar $d_{concave}$, the variation of the light extraction efficiencies was less than 15% for wavelength ranging from 420 nm to 525 nm. The simulation results indicate that the optimum improvement for the LED with PDMS concave microstructures arrays for $\lambda = 420$ nm and $\lambda = 480$ nm occur for $d_{concave} = 0.5\mu$ m. As for the LED emitting at 525nm, the optimum $d_{concave}$ was found as 0.75µm.



Fig. 3. Comparison of light extraction efficiency ratios of InGaN LEDs with varying concave microstructures diameter, and emission wavelength, with PDMS layer thickness of (a) $3.0\mu m$, and (b) $2.0\mu m$.

For comparison purpose, the ray tracing simulation was also conducted for LED devices coated with 2.0µm thick PDMS layer with concave microstructure arrays. Figure 3(b) shows that the concave microstructures arrays lead to increase in light extraction efficiencies by 1.5 to 2.0 times with diameter of the PDMS concave microstructures arrays ($d_{concave}$) ranging from 0.3µm up to 2µm. The simulation results indicate that the optimum improvement for the LED with PDMS concave microstructures arrays for $\lambda = 420$ nm and $\lambda = 480$ nm occur for $d_{concave} = 0.3$ µm. As for the LED emitting at 525nm, the optimum $d_{concave}$ was found as 0.75µm.

3. Experimental works and discussions

A monolayer of SiO_2 microsphere arrays was first deposited on a glass slide using the lowcost rapid convective deposition technique [21,22,26]. The schematic of the rapid convective deposition technique is shown in Fig. 4. 10µL of the monosized SiO₂ colloidal suspension was injected to the corner between the deposition blade and substrate. The deposition blade and the glass slide forms a wedge. The deposition blade was then swept across the glass slide with a linear motor. The strategy behind using colloidal self-assembly is to exploit the tendency of monosized sub-micrometer spheres to spontaneously arrange into a close-packed twodimension (2D) crystal. Strong capillary forces at solid/air/water interface induce crystallization of microspheres into a two-dimension array.



Fig. 4. Schematic of rapid convective deposition of SiO₂ microspheres on glass substrate.

To deposit a monolayer SiO_2 microspheres, optimal deposition speed has to be utilized. Above the critical deposition speed, a sub-monolayer of SiO_2 microsphere arrays was obtained, and multilayers of SiO_2 microspheres were deposited for slower deposition speeds. Details of the optimization studies on the monolayer SiO_2 microspheres deposition conditions and deposition speed can be found in reference 22. The confocal laser scanning micrograph (CLSM) of the monolayer SiO_2 microsphere arrays on the glass substrate is shown in Fig. 5.



Fig. 5. Confocal laser scanning microscopy images of monolayer SiO_2 microspheres arrays on glass substrate, used as imprinting template for forming PDMS concave microstructures arrays.

The deposited SiO₂ microsphere arrays described above served as a template for forming the concave microstructures on PDMS via soft-lithography. In our approach to form the concave microstructures on top of the LED structure, the LED wafer was pre-patterned with 3.6 μ m thick photoresist (PR 1813) to cover the p- and n-metal contacts of the LEDs. The pmetal for the LEDs used in this experiment was Ni/Au (20/300nm) with radial fan-out design for current spreading, and the n-metal was Ti/Au (20/300nm). The patterned photoresist will be subsequently used as a lift off mask for PDMS. The photoresist is sufficiently thick to create a discontinuity in the subsequent PDMS layer to enable the lift off process. A 3.0 μ m