

Optimization of Light Extraction Efficiency of III-Nitride LEDs With Self-Assembled Colloidal-Based Microlenses

Yik-Khoon Ee, *Student Member, IEEE*, Pisist Kumnorkaew, Ronald A. Arif, Hua Tong, Hongping Zhao, James F. Gilchrist, and Nelson Tansu, *Member, IEEE*

Abstract—Improvement of light extraction efficiency of InGaN LEDs using colloidal-based SiO₂/polystyrene (PS) microlens arrays was demonstrated. The size effect of the SiO₂ microspheres and the thickness effect of the PS layer on the light extraction efficiency of III-nitride LEDs were studied. The monolayer rapid convective deposition conditions for SiO₂ microspheres were also investigated. Ray tracing simulations show that the use of microlens arrays can lead to increase in light extraction efficiency of InGaN LEDs by 2.64 times. This is consistent with experiments that demonstrated 2.49 times improvement in light extraction utilizing SiO₂/PS microlens arrays. The enhancement in light extraction efficiency is attributed to increase in effective photon escape cone due to SiO₂/PS microlens arrays, and reduced Fresnel reflection within the photon escape cone due to the grading of refractive index change between GaN/SiO₂/PS/air interface.

Index Terms—Colloid, InGaN quantum wells (QWs), LEDs, light extraction efficiency, microlens arrays.

I. INTRODUCTION

THE LED invented in the 1960s [1] has its initial application mainly as indicator lights in electronic products. However, after the first demonstration of viable double-heterostructure blue and green InGaN LED [2], [3], the current LED technology is demonstrating device performance potential for becoming the next generation illumination platform [4].

One of the issues that has continued to remain a challenge since the inception of the LED is increasing light extraction efficiency of LED devices utilizing a low-cost and practical approach. Most of the light generated by the active regions of

the LEDs is trapped in the higher refractive index semiconductor material. The large refractive index difference of GaN ($n = 2.5$) and air ($n = 1$) at the interface results in total internal reflection that leads to low light extraction efficiency. The escape cone in GaN material is only 23.5° with a photon escape probability of only 4%.

Several research groups have adopted different techniques to improve the light extraction efficiency of LEDs. One approach is to roughen the flat emission surface of the LEDs so that more light at the roughened GaN/air interface is able to scatter out from the active region. This has been done using wet etching [5] or photochemical etching [6], but the roughness obtained using these methods was not uniform, and thus, leads to a variation in the improvement of light extraction efficiency of the LEDs across the sample. Besides roughening the top surface emission area of the LEDs, some research groups have tried to roughen the mesa sidewalls of the LEDs using photochemical etching [7]. Similarly, the surface of the mesa sidewalls was nonuniform. Another approach to increase the light extraction efficiency of the LEDs was to use an oblique mesa sidewall [8]. This was achieved using reflowed photoresist and by adjusting the flow of CF₄ gas during the dry etching process. The improvement of the light extraction efficiency was isolated to the areas in the sidewall regions only.

Photonic crystal structures have also been utilized to enhance the light extraction efficiency of LEDs [9]–[12]. The photonic crystal structures were fabricated using electron beam, laser holographic, or colloidal lithography. Other approaches include fabricating sapphire microlenses [13], nanopillars [14], and graded refractive indexes between GaN/air interface with planar materials [15]–[17]. Recently, we demonstrated a novel approach to significantly enhance the light extraction efficiency of III-nitride LEDs by fabricating SiO₂/polystyrene (PS) microlens arrays [18], [19].

In this paper, we perform detailed studies on the conditions for monolayer SiO₂ deposition on GaN surface, and parametric studies to optimize the light extraction efficiency of InGaN quantum wells (QWs) LEDs employing these SiO₂/PS microlens arrays. The parameters are: 1) diameter of SiO₂ microspheres (d_{SiO_2}) and 2) thickness of the planarized PS layer (h_{PS}). These studies were carried out for optimizing the light extraction efficiency of LEDs with microlens arrays with two configurations: 1) SiO₂/PS microlens arrays and 2) SiO₂ microspheres arrays. The experimental and simulation results are compared with those of planar LEDs.

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Y.-K. Ee, H. Tong, H. Zhao, and N. Tansu are with the Center for Optical Technologies, Department of Electrical and Computer Engineering, Lehigh University, Bethlehem, PA 18015 USA (e-mail: ee@lehigh.edu; hut3@lehigh.edu; hoz207@lehigh.edu; tansu@lehigh.edu).

R. A. Arif was with the Center for Optical Technologies, Department of Electrical and Computer Engineering, Lehigh University, Bethlehem, PA 18015 USA. He is now with EpiWorks, Inc., Champaign, IL 61822 USA.

P. Kumnorkaew and J. F. Gilchrist are with the Center for Advanced Materials and Nanotechnology, Department of Chemical Engineering, Lehigh University, Bethlehem, PA 18015 USA (e-mail: pik205@lehigh.edu; gilchrist@lehigh.edu).

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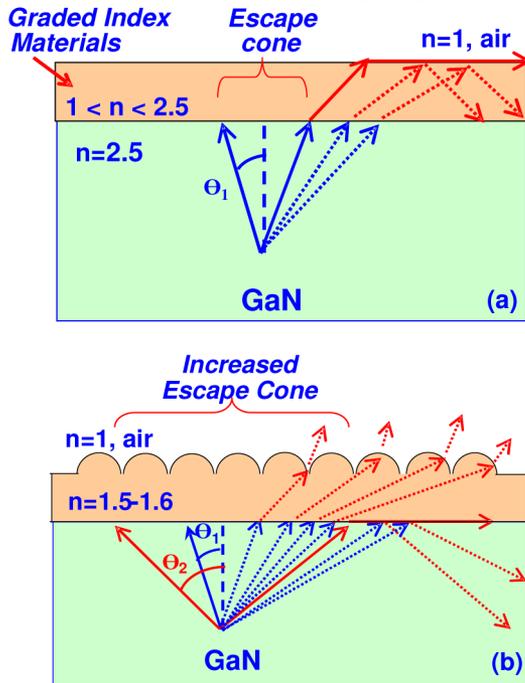


Fig. 1. (a) Schematic of LEDs coated with a planar layer of intermediate refractive index material between the semiconductor/air interface, with no increase in photon escape cone. (b) Schematic of LEDs coated with SiO_2/PS microlens array, with increase in photon escape cone.

II. CONCEPT OF MICROLENS ARRAY USING SiO_2 MICROSPHERES

The large refractive index difference of GaN ($n = 2.5$)/air ($n = 1$) interface results in total internal reflection, which leads to low light extraction efficiency. The total internal reflection leads to narrow escape cone of only 23.5° with an escape probability of only 4% from top surface of the LED due to the optical modes being trapped inside the semiconductor. The light extraction efficiency of the LED can be increased by reducing the Fresnel reflection for the light emitted within the escape cone. This can be done by grading the refractive index change between GaN and air with planar materials. However, the photon escape cone remains unchanged at 23.5° for GaN/air interface, with the rest of the light trapped inside the semiconductor, as shown in Fig. 1(a).

To increase the photon escape cone, SiO_2/PS microlens array was deposited on the emission surface of the LEDs, as shown in Fig. 1(b). The SiO_2 microspheres were semiburied in the PS, forming close-packed lens-like arrays. These arrays allow photons emanating from the QW to be scattered out of the LED structure with larger photon escape cone from 23.5° to 39.8° . The Fresnel reflection was also reduced using the SiO_2/PS microlens arrays. The refractive index of GaN in the visible spectrum is 2.5, while the refractive indexes of PS and SiO_2 microspheres are 1.58 and 1.46, respectively. This provides a graded refractive index change from the GaN/PS/ SiO_2 /air interface, thus reducing Fresnel reflection. Using the SiO_2/PS microlens arrays, the light extraction efficiency of the LEDs can

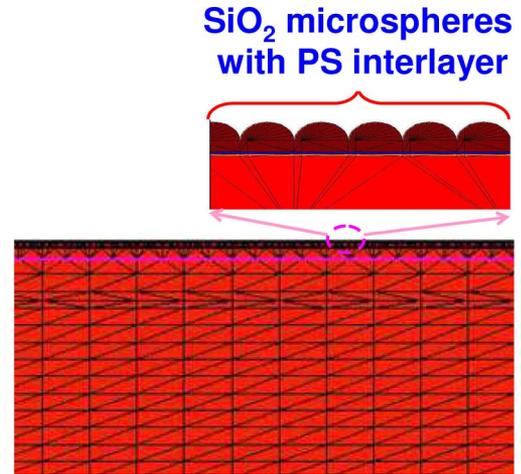


Fig. 2. Schematic of InGaN QWs LEDs simulation mesh structure utilizing SiO_2/PS microlens array on the top emission surface.

be increased with larger photon escape cone and reduction in Fresnel reflection.

III. RAY TRACING SIMULATION OF LIGHT EXTRACTION EFFICIENCY

To study the effect of SiO_2/PS microlens arrays on the light extraction efficiency of the LEDs, a ray tracing simulation was conducted. Monte Carlo ray tracing was used to calculate the light extraction efficiency of the InGaN QWs LED. The Monte Carlo technique was used to simulate the optics design due to the relatively large dimensions of the LED. The simulated LED device size was $100 \mu\text{m} \times 100 \mu\text{m}$. The simulated LED structure includes $400 \mu\text{m}$ sapphire substrate, $2.5 \mu\text{m}$ n-doped GaN, an active region consisting of four-period 2.5 nm InGaN/GaN QWs, and capped with p-doped GaN as the contact layer. The spontaneous and amplified spontaneous emissions of the InGaN QWs were computed and used as the light source for the ray tracing simulation. The interaction of the ray traced photons and carrier transport was carried out in three dimensions self-consistently, taking photon recycling recombination into account. The active mesh domain consists of 74 000 vertexes with 20 rays in each vertex. A total of 1.48 million light rays were used in the simulations, so as to minimize statistical error to less than 0.1%. The schematic of the LED simulation mesh structure utilizing the SiO_2/PS microlens array is shown in Fig. 2.

The flow chart of the Monte Carlo simulation is shown in Fig. 3. First, the physical LED structure was defined, together with the input material parameters such as refractive index and absorption coefficients. Additional LED ray tracing parameters such as number of rays per vertex and minimum intensity of ray before ray tracing terminates were also defined. Ray tracing of the photons starts with the spontaneous emission from the active region, which comprises of a four-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

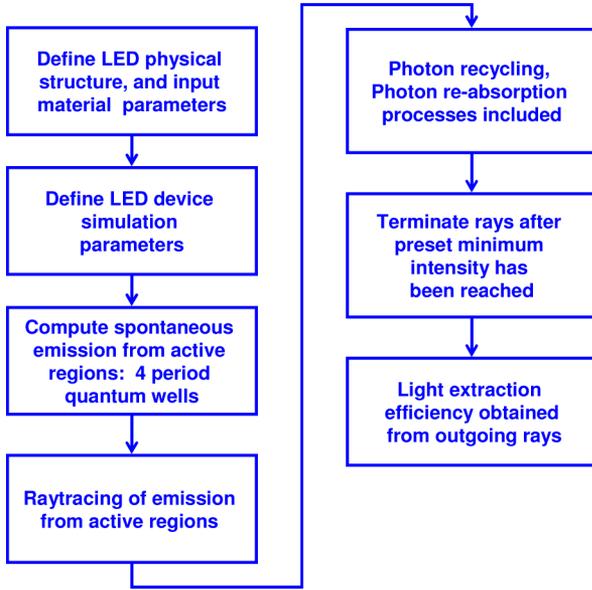


Fig. 3. Flow chart of Monte Carlo ray tracing simulation to calculate the light extraction efficiency of the LED.

of the light extraction efficiency of the III-nitride LED employing microlens arrays with that of planar LED is compared for various SiO_2 microsphere diameters and PS thicknesses with various emission wavelengths.

To investigate the size effect of SiO_2 microspheres on the light extraction efficiency of III-nitride LEDs, ray tracing simulation was performed for an LED with the top contact region coated with SiO_2 microspheres only (no PS layer). The ratio of the light extraction efficiencies of the LED with SiO_2 microspheres compared to those of the planar LED is shown in Fig. 4. The simulation was conducted for three emission wavelengths: 420, 480, and 525 nm. The diameter of the SiO_2 microspheres ranged from 0.0 (planar case) to 3 μm . From the simulation, the use of SiO_2 microspheres alone leads to enhancement of the light extraction efficiency by as high as 1.8 times. When the size of the SiO_2 microspheres increases ($d_{\text{SiO}_2} > 2 \mu\text{m}$), the light extraction efficiency enhancement reduces.

The light extraction efficiency for the LED coated with microlens arrays was also investigated. Fig. 5 shows the ratio of the light extraction efficiencies of the LEDs with microlens arrays compared to those of the planar LED. In this simulation, the SiO_2 microspheres are semiburied in a layer of PS in the emission area of the LED. The thickness of the PS layer is equal to half the diameter of the SiO_2 microspheres. The effect of emission wavelength and the size effect of the SiO_2 microspheres on the light extraction efficiency of LEDs were investigated. Simulations were conducted for LEDs emission at 420, 480, and 525 nm, with diameters (d_{SiO_2}) ranging from 0.5 to 2 μm . From the simulations, the use of these microlens arrays leads to enhancement in the light extraction efficiencies from 1.8 up to 2.7 times for d_{SiO_2} ranging from 0.5 up to 2 μm . The light extraction efficiency enhancement using SiO_2 microspheres is relatively independent of the three different emission wavelengths. For LEDs with similar d_{SiO_2} , the vari-

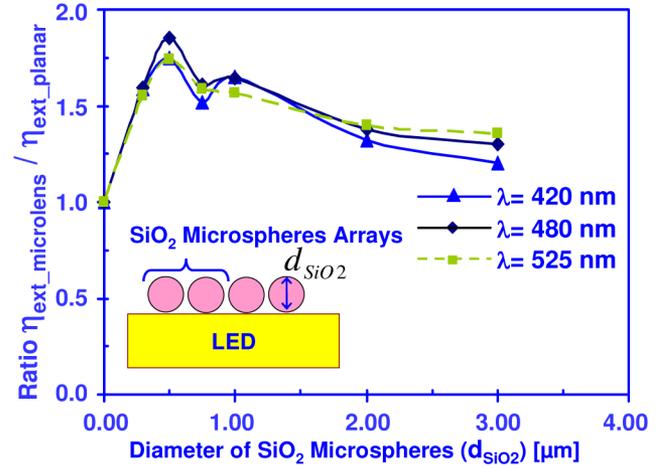


Fig. 4. Comparison of light extraction efficiency ratios of InGaN LEDs with varying SiO_2 diameter (no PS), and emission wavelength.

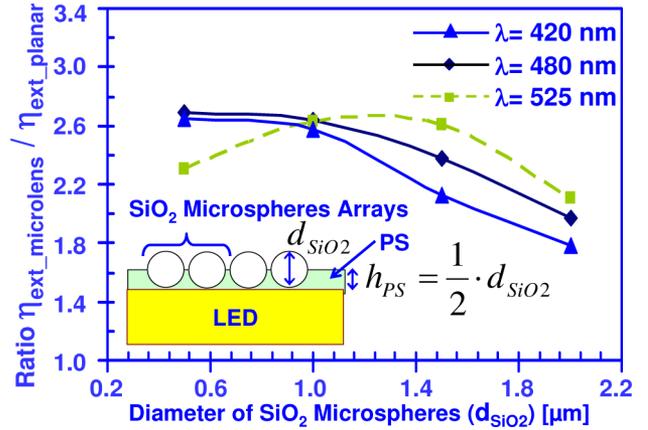


Fig. 5. Comparison of light extraction efficiency ratios of InGaN LEDs with varying SiO_2 diameter and emission wavelength.

ation of the light extraction efficiencies was less than 15% for wavelength ranging from 420 to 525 nm. Simulation indicates that the optimum light extraction efficiency improvement for LED with microlens with wavelength emission of 420 and 480 nm occurred when $d_{\text{SiO}_2} = 0.5 \mu\text{m}$. As for the LED emitting at 525 nm, the optimum d_{SiO_2} was found as 1.25 μm .

Simulations were also conducted to investigate the effect of the PS thickness on the light extraction efficiency of the InGaN QWs LEDs with SiO_2/PS microlens arrays, and are shown in Fig. 6(a) and (b). In the analysis in Fig. 6(a), SiO_2 microspheres with $d_{\text{SiO}_2} = 0.5 \mu\text{m}$ were employed, while varying the thicknesses of the PS layer from 0.0 (no PS layer) up to 0.35 μm . As shown in Fig. 6(a), it was found that the light extraction efficiency for the LED with $d_{\text{SiO}_2} = 0.5 \mu\text{m}$ microlens arrays was optimized for the case $h_{\text{PS}} = 0.25 \mu\text{m}$, with improvement up to 2.7 times for the case of 480 nm emitting LED. When the PS layer is thicker than the optimum thickness of $h_{\text{PS}} = 0.25 \mu\text{m}$, the effective photon escape cone is reduced as less surface of the SiO_2 microspheres is exposed. On the contrary, when the PS layer is thinner than the optimum thickness, less photons

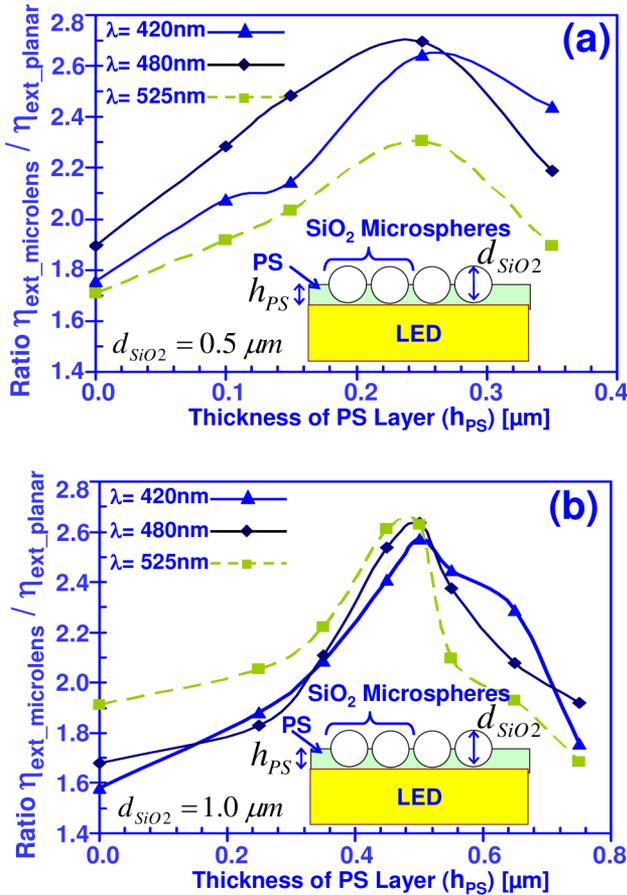


Fig. 6. Comparison of light extraction efficiency ratios of InGaN LEDs with varying PS thickness and emission wavelengths using (a) $d_{\text{SiO}_2} = 0.5\ \mu\text{m}$ and (b) $d_{\text{SiO}_2} = 1.0\ \mu\text{m}$.

are coupled into the SiO₂ microspheres for light extraction. Hence, the maximum effective photon escape cone is achieved, when the optimum PS layer thickness is used, as it allows for the optimum amount of photons to be coupled into the SiO₂ microspheres.

For the analysis in Fig. 6(b), SiO₂ microspheres with $d_{\text{SiO}_2} = 1.0\ \mu\text{m}$ were utilized, while varying the thicknesses of the PS layer from 0.0 (no PS layer) up to 0.75 μm . Similarly, the light extraction efficiency for the LED with $d_{\text{SiO}_2} = 1.0\ \mu\text{m}$ microlens arrays was optimized for the case $h_{\text{PS}} = 0.5\ \mu\text{m}$ ($d_{\text{SiO}_2} = 1.0\ \mu\text{m}$), with improvement up to 2.6 times for the case of 525 nm emitting LED, as shown in Fig. 6(b). Hence, the light extraction efficiency of the LEDs are optimized when the thickness of the PS layer is half that of the diameter of the SiO₂ microspheres [$h_{\text{PS}} = (1/2) \times d_{\text{SiO}_2}$].

IV. DEPOSITION PROCESS OF SiO₂/PS MICROLENS

The *ex situ* rapid convective deposition [19], [20] of the microsphere layers from colloidal suspensions were subsequently conducted on top of the InGaN QW photoluminescence (PL) and LEDs samples. A schematic of the rapid convective deposition technique is shown in Fig. 7. The strategy behind using colloidal self-assembly is to exploit the tendency of monosized

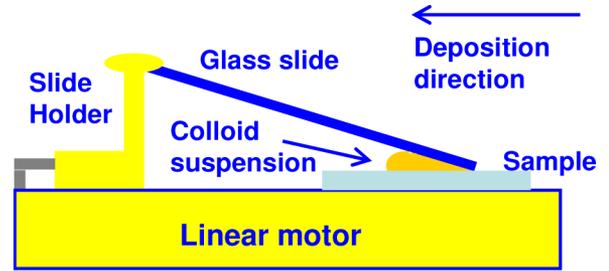


Fig. 7. Schematic of rapid convective deposition of PS and SiO₂ microspheres on LED samples.

submicrometer spheres to spontaneously arrange into a close-packed 2-D crystal via strong capillary forces.

To optimize the deposition of monolayer SiO₂ microspheres, an optimal deposition speed was utilized. If the deposition speed was too fast, a submonolayer of SiO₂ microspheres was obtained. If the deposition speed was too slow, then multilayer SiO₂ microsphere layers were obtained. Only when the deposition speed was optimized, a monolayer SiO₂ microsphere array was deposited. The confocal laser scanning micrographs of the three different morphologies are shown in Fig. 8.

The deposition blade can be made hydrophilic or hydrophobic, and the deposition angle of the blade can also be varied. The optimal deposition conditions and deposition speed to obtain monolayer SiO₂ microspheres have been investigated in [18]. For the monolayer deposition of 0.5 μm SiO₂ microspheres and 1.0 μm SiO₂ microspheres, the optimal deposition speed using a hydrophilic blade at an inclination angle of 80° should be 65 and 60 $\mu\text{m}/\text{s}$, respectively.

To create SiO₂/PS microlens arrays (Fig. 9), first a monolayer of hexagonal close-packed PS microspheres was deposited followed by a monolayer of SiO₂ microspheres. The sample was then heated to 140 °C to planarize the PS, semiburying the SiO₂ microspheres forming convex lens-like structures on top of the light emission areas of the PL and LEDs samples. The capturing of the SiO₂ microspheres in the planar PS occurred without significant rearrangement to the close-packed structure of the microspheres.

V. EXPERIMENTAL RESULTS

The InGaN QWs PL samples for experiments with and without the SiO₂/PS microlens arrays were grown at the same time in the metal-organic chemical vapor deposition (MOCVD) reactor. Room temperature PL measurements were done using a 325-nm laser as the excitation source from the backside of the samples, with the PL luminescence collected from the top of the samples. The PL samples were coated with the following configurations: 1) SiO₂ microspheres (no PS layer) in one region and 2) SiO₂/PS microlens arrays in another region. The PL luminescence of the sample emitting at 429 nm with and without coatings is shown in Fig. 10. There is a 10 nm variation in the peak PL luminescence of the different coated regions. This PL spatial inhomogeneity is due to variation of indium content of the InGaN QWs across the wafer [21]. As shown in Fig. 10, the

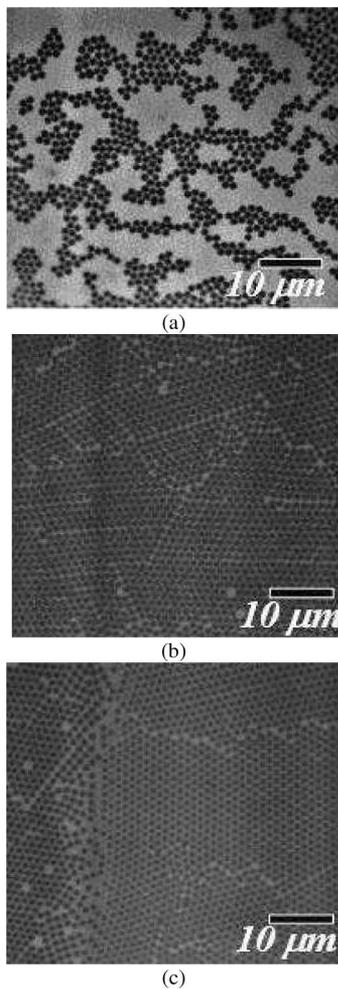


Fig. 8. Confocal laser scanning microscopy images of (a) submonolayer SiO_2 microspheres deposition when the deposition speed was too fast, (b) monolayer SiO_2 microspheres when deposition speed was optimized, and (c) multilayer SiO_2 microspheres deposition when the deposition speed was too slow. [19].

integrated PL luminescence of the sample covered with $0.5 \mu\text{m}$ SiO_2 microspheres and $0.5 \mu\text{m}$ $\text{SiO}_2/0.1 \mu\text{m}$ PS microlens arrays exhibited enhancement of 1.69 and 2.00 times over that of the uncoated sample, respectively. These are in good agreement with the simulation, which predicts an enhancement of 1.75 and 2.08 times for the InGaN QWs coated with $0.5 \mu\text{m}$ SiO_2 microspheres only ($h_{\text{PS}} = 0 \mu\text{m}$) and the $0.5 \mu\text{m}$ $\text{SiO}_2/0.1 \mu\text{m}$ PS microlens arrays, respectively.

PL studies were also conducted for InGaN QWs samples coated with $1.0 \mu\text{m}$ $\text{SiO}_2/h_{\text{PS}}$ of $0.5 \mu\text{m}$ PS microlens arrays. Fig. 11 shows the PL spectra of the coated and uncoated samples. The integrated PL luminescence for samples with the $1.0 \mu\text{m}$ SiO_2/PS microlens arrays showed 2.62 times improvement over that of the uncoated sample. This is in agreement with the simulated ray tracing results that predicted a 2.57 times improvement in light extraction efficiency for the case of $1.0 \mu\text{m}$ SiO_2/PS microlens arrays.

To investigate the effect of light extraction efficiency enhancement with longer emission wavelength, PL studies were also conducted on $\text{In}_{0.22}\text{Ga}_{0.78}\text{N}$ QWs samples with emis-

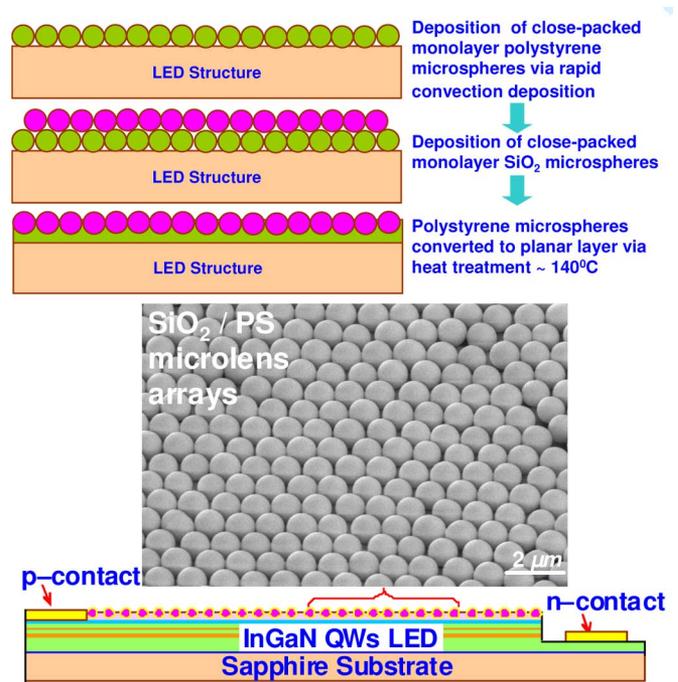


Fig. 9. Process flow schematic of depositing SiO_2/PS microlens array on PL and LEDs samples using rapid convective deposition technique, and schematic of InGaN QWs LEDs structure utilizing SiO_2/PS microlens array, with scanning electron microscopy image showing SiO_2/PS in 2-D hexagonal close-packed microlens array.

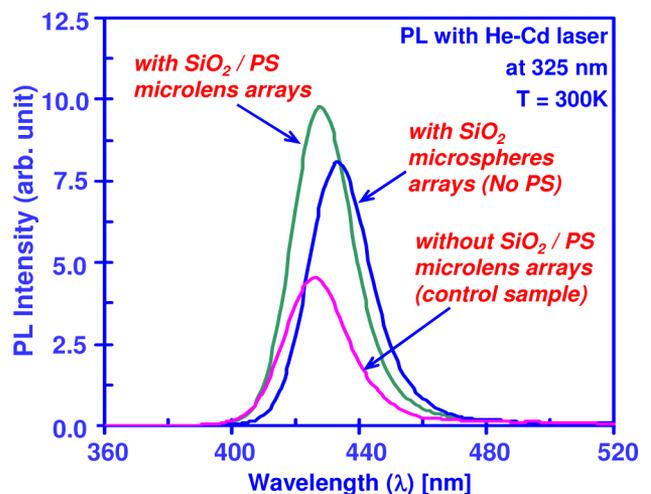


Fig. 10. Comparison of photoluminescence intensity of $\text{In}_{0.12}\text{Ga}_{0.88}\text{N}$ QWs sample coated with $0.5 \mu\text{m}$ SiO_2/PS microlens array, $0.5 \mu\text{m}$ SiO_2 microspheres alone, and uncoated sample.

sion in the green regime at $\lambda_{\text{peak}} = 517 \text{ nm}$. Fig. 12 shows the PL spectra of the $\text{In}_{0.22}\text{Ga}_{0.78}\text{N}$ QW PL samples ($\lambda_{\text{peak}} = 517 \text{ nm}$) without any coating, and with the $0.5 \mu\text{m}$ $\text{SiO}_2/0.1 \mu\text{m}$ PS microlens arrays. The integrated PL luminescence for samples with the SiO_2/PS microlens arrays showed 1.84 times improvement over that of the uncoated sample. This is in good agreement with the simulated ray tracing results, which

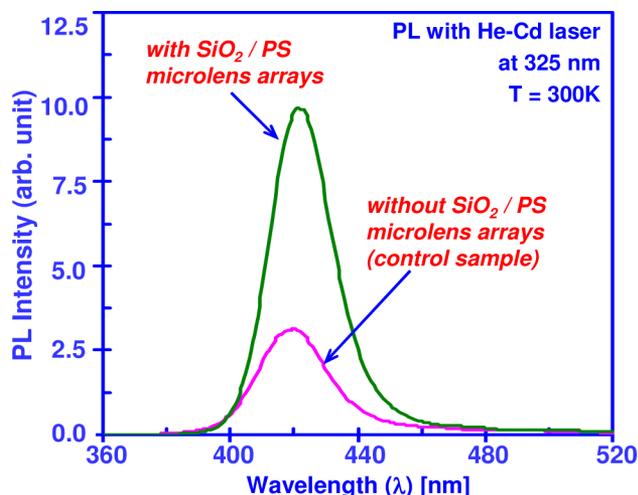


Fig. 11. Comparison of photoluminescence intensity of InGaN QWs sample coated with $1.0 \mu\text{m}$ SiO_2/PS microlens array and uncoated sample.

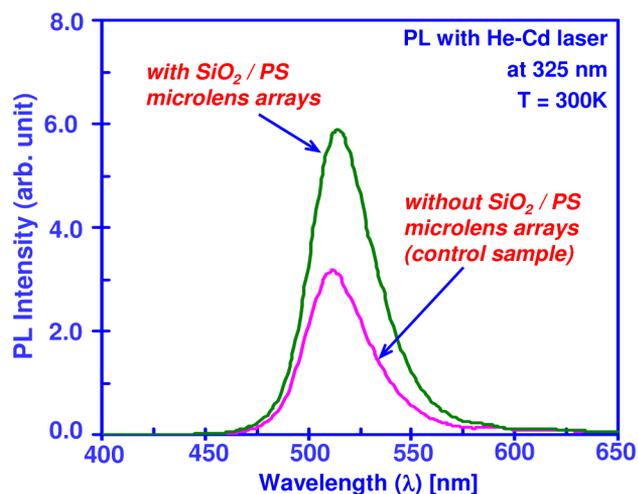


Fig. 12. Comparison of photoluminescence intensity of $\text{In}_{0.22}\text{Ga}_{0.78}\text{N}$ QWs sample coated with $0.5 \mu\text{m}$ $\text{SiO}_2/0.1 \mu\text{m}$ PS microlens array and uncoated sample.

predicted a 1.91 times improvement in light extraction efficiency for the case of $0.5 \mu\text{m}$ $\text{SiO}_2/0.1 \mu\text{m}$ PS microlens arrays.

The SiO_2/PS microlens arrays were also coated on InGaN LEDs devices. The wafer-level optical output power of a four-period $\text{In}_{0.19}\text{Ga}_{0.81}\text{N}$ MQWs LED with injection current ranging from 0 to 100 mA is shown in Fig. 13. The LED devices were measured under continuous-wave condition at room temperature. The on-wafer output power of the LEDs with an area of 1 mm^2 were measured in a dark room, with driving current up to 100 mA for both coated and uncoated LEDs. The LED coated with $1.0 \mu\text{m}$ $\text{SiO}_2/0.5 \mu\text{m}$ PS microlens arrays exhibited a 2.49 times improvement in the output power as compared to that of the planar uncoated LED at a current injection level of 100 mA. This improvement is in good agreement with simulated enhancement of 2.64 times.

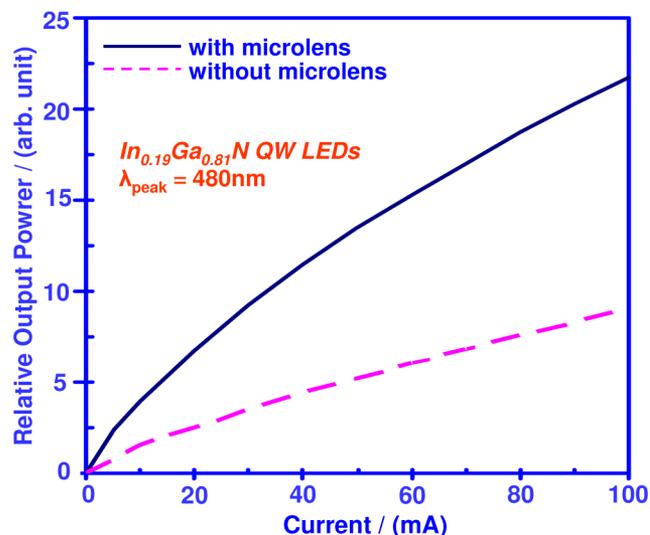


Fig. 13. Comparison of electroluminescence intensity of InGaN QW LEDs emitting in 480 nm region, with and without SiO_2/PS microlens array.

VI. CONCLUSION

From our studies, we found that the use of SiO_2/PS microlens arrays on top of III-nitride LED device leads to a low cost and practical approach to increase its light extraction efficiency. Comprehensive studies have been conducted to optimize the light extraction efficiency of III-nitride LEDs with SiO_2/PS microlens arrays. The size effects of the SiO_2 microspheres and the influence of the thickness of the PS layer on the light extraction efficiency of the LEDs have been investigated. From our simulation studies, the use of SiO_2 microsphere arrays on top of III-nitride LEDs leads to significant increase of the light extraction efficiency by 1.8 times in comparison to those of planar LEDs for emission wavelengths of 420–525 nm. The simulation studies indicated that the use of SiO_2/PS microlens arrays led to improvement of light extraction efficiency in the range of 1.8 up to 2.7 times, depending on the thickness of the PS layer. For the case of III-nitride LEDs employing only SiO_2 microspheres, the improvement is relatively independent of emission wavelength. For the III-nitride LEDs employing SiO_2/PS microlens arrays, the increase in the light extraction efficiency depends strongly on the diameter of SiO_2 microspheres (d_{SiO_2}) and thickness of PS layer (h_{PS}). From our studies, we found that the optimum d_{SiO_2} for 420–525 nm emitting LEDs ranges from 0.5 up to $1.25 \mu\text{m}$. The optimum thickness of the PS layer in the SiO_2/PS microlens arrays configuration is found as half of the diameter of SiO_2 microspheres (d_{SiO_2}).

The experiments were also carried out for III-nitride LEDs employing SiO_2 microspheres only and SiO_2/PS microlens arrays configurations. The deposition technique was performed employing rapid convective deposition, and the details of the deposition process were described in [17] and [18]. Optimized volume fraction and deposition speed are very important parameters to ensure microsphere monolayer deposition, rather than submonolayer or multilayers microsphere depositions. The use of $0.5\text{-}\mu\text{m}$ -diameter SiO_2 microspheres and $0.5 \mu\text{m}$ $\text{SiO}_2/0.1 \mu\text{m}$

PS microlens arrays on top of InGaN QWs PL sample led to improvement in the light extraction efficiency by 1.69 and 2.00 times, respectively, in comparison to those measured from planar LED samples. The experimental results are in good agreement with the Monte Carlo ray tracing simulation that predicts light extraction efficiency improvement of 1.75 and 2.08 times in those two respective cases.

The SiO₂/PS microlens arrays were also deposited on InGaN QWs LEDs emitting in 480 nm spectral range. The use of 1.0- μ m-diameter SiO₂/0.5 μ m PS microlens arrays on the LEDs led to a 2.49 times improvement in the light extraction efficiency in comparison with that of planar LEDs. This enhancement is in good agreement with the simulation, which predicted an enhancement of 2.64 times.

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Yik-Khoon Ee received the Bachelor's degree in electrical and electronic engineering (first-class honors) from Nanyang Technological University, Singapore, in 2003, the M.S. degree in 2007 from Lehigh University, Bethlehem, PA, where he is currently working toward the Ph.D. degree.

He was with Denselight Semiconductors and Agilent Technologies. He is engaged in the field of semiconductor optoelectronics, metal-organic chemical vapor deposition (MOCVD) epitaxy, device fabrication of III-nitride LEDs, dislocation density reduction of III-nitride material on sapphire, and using novel techniques to increase the light extraction efficiency of LEDs. He has authored or coauthored more than 46 refereed journal and conference publications.

Mr. Y.-K. Ee was the recipient of the Best Student Paper Award from IEEE Photonics Global Conference 2008.

Pisist Kumnorkaew received the Bachelor's and Master's degrees in chemical engineering from King's Mongkut Institute of Technology, Bangkok, Thailand, in 2000 and 2003, respectively. He is currently working toward the Ph.D. degree in chemical engineering at Lehigh University, Bethlehem, PA, on a Royal Thai Scholarship.

He was a University Lecturer at Mae Fah Laung University. He is engaged in the research on the monolayer deposition of micro- and nanoparticles using rapid convective deposition technique in terms of fundamental physics and applications.

Ronald A. Arif received the Bachelor's degree from Nanyang Technological University, Singapore, in 2002, and the Master's degree and the Ph.D. degree in electrical engineering from Lehigh University, Bethlehem, PA, in 2005 and 2008, respectively.

From 2002 to 2003, he was a Process Engineer with Agilent Technologies, Singapore. He is currently a Member of Technical Staff at EpiWorks, Inc., Champaign, IL. He is engaged in fundamental studies and novel approaches to improve radiative efficiency of visible gain media based on III-nitride semiconductor nanostructures for high-efficiency LEDs and lasers, in particular for solid-state lighting. He has authored or coauthored more than 45 refereed journal and conference publications.

Dr. Arif was a recipient of Sherman Fairchild Fellowship on Solid State Studies at Lehigh University.

Hua Tong received the B.S. and M.S. degrees in electronics engineering from Tsinghua University, Beijing, China, in 1998 and 2001, respectively. He is currently working toward the Ph.D. degree at the Department of Electrical and Computer Engineering (ECE), Lehigh University, Bethlehem, PA.

He is engaged in the theoretical/computational analysis of the physics, epitaxy [metal–organic chemical vapor deposition (MOCVD)] and fabrication of semiconductor devices. His current research interests include fundamental studies of thermoelectric properties of III-nitride material, MOCVD epitaxy, and fabrications of InGaN thermoelectric devices on GaN substrates.

Hongping Zhao received the Bachelor's degree in physics from Nanjing Normal University, Nanjing, China, in 2003, and the Master's degree in electrical engineering from Southeast University, Nanjing, in 2005. She is currently working toward the Ph.D. degree at the Electrical and Computer Engineering Department, Lehigh University, Bethlehem, PA.

She is engaged in research in the areas of device physics, epitaxial growth, and fabrication of semiconductor optoelectronics devices based on semiconductor nanostructures. Her current research interests include fundamental studies and approaches to improve gain and spontaneous emission of visible gain media based on III-nitride semiconductors for high-performance lasers, and metal–organic chemical vapor deposition (MOCVD) epitaxy of InN material for quantum dots and solar cell applications. She has authored or coauthored more than 40 refereed journal and conference publications.

Ms. Zhao is the recipient of the 2008 International Society of Optical Engineers (SPIE) Educational Scholarship and Sherman Fairchild Fellowship on Solid State Studies.

James F. Gilchrist received the B.S. degree in chemical engineering from Washington University, St. Louis, MO, in 1997, and the Ph.D. degree from Northwestern University, Evanston, IL, in 2003.

He was a Postdoctoral Research Associate in the Department of Materials Science and Engineering, University of Illinois, Urbana. Since 2004, he has been the P.C. Rossin Assistant Professor of chemical engineering at Lehigh University, Bethlehem, PA, where he is also a Member of the Center of Advanced Materials and Nanotechnology, the Center for Polymer Science and Engineering, an Executive Board Member of the Lehigh Nanotechnology Network, and directs the Laboratory for Particle Mixing and Self-Organization. His current research interests include areas of fluidic and particle technologies including nanoparticle self-assembly, suspension transport, hemodynamics, microfluidics, chaotic mixing, and granular dynamics. He has received funding from the National Science Foundation, the National Aeronautics and Space Administration, the Petroleum Research Fund, and was selected as the 2007 North American Mixing Forum Startup Grantee.

Nelson Tansu (S'99–M'02) was born on October 1977. He received the B.S. degree (with highest distinction) in applied mathematics, electrical engineering, and physics, and the Ph.D. degree in electrical engineering from the University of Wisconsin-Madison, Madison, in May 1998 and May 2003, respectively.

Since July 2003, he has been an Assistant Professor in the Department of Electrical and Computer Engineering (ECE) and the Center for Optical Technologies (COT) at Lehigh University, Bethlehem, PA, where since April 2007, he has been the Peter C. Rossin Assistant Professor of electrical and computer engineering. He is engaged in research on the theoretical and experimental aspects of the physics of semiconductor optoelectronics materials and devices, the physics of low-dimensional semiconductor (nanostructure), and metal–organic chemical vapor deposition (MOCVD) epitaxy, and device fabrications of III-nitride and III–V-nitride semiconductor optoelectronics devices on GaAs, InP, and GaN substrates. His teaching interests are in the areas of optoelectronics and photonics, semiconductor physics, applied quantum mechanics, and engineering electromagnetism. He has authored or coauthored numerous refereed international journal and conference publications (total > 148), and holds several U.S. patents.

Dr. Tansu was a panel member for the U.S. National Science Foundation, the U.S. Department of Defense, and other agencies in U.S. and abroad. He has also given numerous lectures, seminars, and invited talks (total > 35) in universities, research institutions, and conferences in USA, Canada, Europe, and Asia. He is the Primary Guest Editor of the IEEE JOURNAL OF SELECTED TOPICS IN QUANTUM ELECTRONICS Special Issue on Solid State Lighting in 2008–2009. He was an Invited General Participant at the 2008 National Academy of Engineering (NAE)'s U.S. Frontiers of Engineering (FOE) Symposium, and is also the member of the Organizing Committee for the 2009 NAE's U.S. Frontiers of Engineering Symposium. He was a recipient of the Bohn Scholarship, the WARF Graduate University Fellowship, the Vilas Graduate University Fellowship, and the Graduate Dissertator Travel Funding Award, the 2003 Harold A. Peterson ECE Best Research Award (1st Prize) at the University of Wisconsin-Madison, and the 2008 Libsch Early Career Research Award at Lehigh University.