Research Article

Enhanced Optical Output of Near-Ultraviolet Light-Emitting Diodes by a Monolayer of Nanospheres

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Received 30 September 2019; Revised 4 February 2020; Accepted 15 February 2020; Published 20 April 2020

Academic Editor: Sergio Bietti

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The optical output of near-ultraviolet (NUV) light-emitting diodes (LEDs) was improved by including a monolayer of hexagonal close-packed polystyrene (PS) nanospheres. PS nanospheres with different sizes were deposited on the indium tin oxide layer of the NUV LEDs. The electroluminescence results showed that the light extraction efficiency of the NUV LEDs was increased by the inclusion of PS nanospheres, and the maximum optical output enhancement was obtained when the size of the nanospheres was close to the light wavelength. The largest enhancement of the optical output of 1.27-fold was obtained at an injection current of 100 mA. The enhanced optical output was attributed to part of the incident light beyond the critical angle being extracted when the exit surface of the NUV LEDs had a PS nanosphere monolayer. This method may serve as a low-cost and effective approach to raise the efficiency of NUV LEDs.

1. Introduction

Near-ultraviolet (NUV) light-emitting diodes (LEDs) have been used in various applications such as biological agent identification, ultraviolet curing, fake bill detection, and solid-state lighting [1–3]. Although gallium nitride- (GaN-) based NUV LEDs are now being produced commercially, their capacities still need to be improved. For example, the external quantum efficiency (EQE) of NUV LEDs is still unsatisfactorily low [4]. One of the main causes of the low EQE of NUV LEDs is their low light extraction efficiency (LEE), which is caused by the total internal reflection by the refractive index difference between the semiconductor and air [5]. Various approaches have been used to enhance the LEE of NUV LEDs, such as surface plasmon enhancement [6–8], surface roughening [9–11], aluminum (Al) n-electrodes [12], periodic hole patterns [13], additional silver nanostructures [14–17], and Al-doped indium tin oxide (ITO) electrodes [18].

Arrays of periodic nanospheres exhibit unique scattering properties because of the Mie resonance of individual spheres

and Bragg diffraction arising from the periodic arrangement of the nanospheres [19]. Various types of nanospheres have been used to enhance the LEE of blue LEDs, and the outstanding enhancement has been achieved [20–27]. This approach has advantages of low cost, high throughput, and large area [21]. Additionally, this approach can avoid degrading the electrical properties of the devices because it does not involve etching or lithographic processing [27]. However, the design and fabrication of nanospheres on the ITO layer of commercial NUV LEDs have not been studied. It is very important to determine the optimal size of nanospheres for specific-wavelength NUV LEDs.

In this paper, we study the optical output enhancement of a monolayer of hexagonal close-packed polystyrene (PS) nanospheres adsorbed on the ITO layer of NUV LEDs. To investigate the effect of PS nanosphere size on the optical properties of the NUV LEDs, monolayers of nanospheres with different sizes are prepared on the NUV LEDs. PS nanospheres with diameters of 250, 300, 400, 600, 800, 1000, 1200, and 1500 nm are investigated. The electroluminescence results show that a PS nanosphere monolayer increases the

optical output compared to that of the corresponding LED without PS nanospheres. The maximum optical output enhancement is achieved using PS nanospheres with a diameter of 400 nm. The light output power of the LEDs with a monolayer of 400 nm PS nanospheres increased by 127% at an injection current of 100 mA compared with that of the corresponding LED without a nanosphere monolayer. The experimental results are confirmed by a three-dimensional (3D) finite difference time-domain (FDTD) simulation. The improvement of NUV LED performance and the physical mechanism of light extraction are discussed in detail.

2. Experimental Methods

Figure 1 shows a schematic diagram of an NUV LED with a monolayer of hexagonal close-packed PS nanospheres. The planar NUV (385 nm) AlGaIn/InGaIn multiple quantum well (MQW) LED structure was grown on a 2-inch sapphire substrate by metal–organic chemical vapor deposition. The LED structures consisted of a sapphire substrate, 2 μm thick undoped GaN layer, 3 μm thick n-GaN layer, 300 nm thick five-period AlGaIn/InGaIn MQW active layer, and 100 nm thick p-GaN layer. The device surface was a transparent ITO electrode, which acted as a current spreading layer. LEDs with dimensions of $185 \times 275 \mu\text{m}$ were formed by mesa etching of the exposed n-type GaN via standard photolithography, ITO wet etching, and subsequent inductively coupled plasma etching. Cr/Pt/Au was deposited on top of the ITO surface as well as the exposed n-GaN layer as contacts for both the p-GaN and n-GaN layers. Nanosphere monolayer arrays were formed on top of the ITO surface of NUV LEDs by self-assembly [28, 29]. To study the influence of nanosphere size on the LEE of NUV LEDs, we fabricated monolayers of PS nanospheres with different diameters on the ITO surface of the LEDs. All of the devices were fabricated from the same LED wafer to eliminate the variations from wafer to wafer and ensure the reliability of the results. The light output power of the LED chip was measured using an integrating sphere.

3. Results and Discussion

Figure 2 shows scanning electron microscopy (SEM) images of monolayers of PS nanospheres with different sizes. The size and distribution of PS nanospheres were relatively uniform. Apart from a few defects, the PS nanospheres were closely arranged in a single layer to form a hexagonal close-packed periodic structure. In experiments, a 2-inch LED wafer was divided into eight parts to prepare the samples. Sample A was a conventional LED without a PS nanosphere monolayer, which acted as a reference device. Samples B, C, D, E, F, G, H, and I were coated with monolayers of PS nanospheres with diameters of 250, 300, 400, 600, 800, 1000, 1200, and 1500 nm, respectively. The morphologies of the PS nanosphere monolayers in samples B, C, D, E, F, G, H, and I are shown in Figures 2(a)–2(h), respectively. The PS nanospheres strongly adhered on the ITO surface.

Figure 3(a) illustrates the injection current–light output ($I-L$) characteristics of NUV LEDs with and without a

FIGURE 1: Schematic diagram of the structure of an NUV LED with a monolayer of hexagonal close-packed PS nanospheres.

monolayer of hexagonal close-packed PS nanospheres. The optical output power at the same injection current increased upon including a monolayer of PS nanospheres. The optical output power of samples B, C, D, E, F, G, H, and I increased by 118%, 123%, 127%, 121%, 119%, 120%, 114%, and 110%, respectively, compared with that of the reference sample A at an I of 100 mA. Figure 3(b) shows the injection current–voltage ($I-V$) characteristics of the NUV LEDs with and without PS nanosphere monolayers measured at room temperature. The forward voltages of the NUV LEDs with and without PS nanosphere monolayers were almost same (3.3 V at 20 mA), and all the $I-V$ curves almost overlapped. Therefore, the deposition of a PS nanosphere monolayer on the LED surface did not markedly degrade the electrical properties of the NUV LEDs.

To verify the experimental results, a 3D FDTD simulation was carried out to investigate the size effect of a hexagonal close-packed PS nanosphere monolayer on the light extraction behavior of the NUV LEDs. The simulated LED structure is shown in Figure 4(a), which consisted of a 1000 nm sapphire substrate, 5200 nm GaN layer (including n-GaN, MQW, and p-GaN layers), and 300 nm ITO layer. In the simulation, a dipole as a point light source was used and the wavelength was 385 nm, which corresponded to the center wavelength of the emission spectrum. The refractive indices of GaN, ITO, and PS are approximately 2.745, 1.9, and 1.59, respectively, at a wavelength of 385 nm [20, 30]. Monolayers of hexagonal close-packed PS nanospheres with different sizes between 250 and 1500 nm were investigated in the simulation. In the simulation, perfectly matched layers (PML) absorbing boundary conditions were assumed on all sides of the model. There were about 16×17 nanospheres in the simulation region. Figure 4(b) shows the calculated and experimental light extraction enhancement ratio of the LEDs as a function of the size of the PS nanospheres. The enhancement ratio is defined as the optical output power of the LEDs coated with monolayers divided by the conventional LED. The trend of the calculated results was consistent with that of the experimental results. The simulated ratios were lower than those determined experimentally, which may be because a plane detector was used in the simulation and an integrating sphere is used in the experiments. And the integrating sphere can collect more light. Both the

FIGURE 2: SEM images of monolayers of PS nanospheres with sizes of (a) 250 nm, (b) 300 nm, (c) 400 nm, (d) 600 nm, (e) 800 nm, (f) 1000 nm, (g) 1200 nm, and (h) 1500 nm.

experimental and calculated results showed that the optimum extraction efficiency was achieved when the scattering particles had a size close to the wavelength of light, which is consistent with the results in Refs. [27, 31].

The above results show that the formation of a monolayer of hexagonal close-packed PS nanospheres on the ITO surface of LEDs can improve their LEE. To investigate the physical mechanism of the LEE enhancement, the transmission spectrum of the related structures at an emission wavelength of 385 nm was calculated by FDTD simulation; the results obtained for s and p polarization are shown in Figures 5(a) and 5(b), respectively. For the conventional LEDs without a PS nanosphere monolayer, the transmission decreased to zero beyond the critical angle of 21.5° . After

being covered with a monolayer of 400 nm PS nanospheres, two extra transmission peaks appeared at about 23° and 31° for both polarizations. This implies that the incident light beyond the critical angle can be partly extracted when the LED is coated with PS nanospheres, leading to enhanced LEE.

It is important to distinguish the physical mechanisms that led to the appearance of the two additional peaks in the transmission spectra. We calculated spatial distributions of electric field intensity of three adjacent spheres corresponding to the two peaks for p polarization, as shown in Figures 5(c) and 5(d). In the simulation, the Bloch boundary conditions were set around the unit cell and PML absorbing boundary conditions were used at the top and bottom boundaries of the cell. The distribution of electric field

FIGURE 3: Electroluminescence curves of the NUV LEDs with and without PS nanosphere monolayers. (a) Optical output power versus injection current and (b) injection current versus voltage characteristics.

FIGURE 4: (a) Schematic illustration of the LED simulation model. (b) Dependence of the experimental and simulated enhancement ratios of light extraction at an emission wavelength of 385 nm on PS nanosphere diameter.

intensity with an incident angle of 23° showed that the light could be propagated to air through the PS nanospheres. Therefore, it is generally believed that the transmission peak at about 23° originates from the diffraction of the evanescent field near the PS-ITO interface caused by the guide modes, which is similar to the light extraction behavior of a typical two-dimensional (2D) photonic crystal [19, 32]. However, the distribution of electric field intensity with an incident angle of 31° shows that the light can be extracted to a PS nanosphere and then exhibits strong confinement inside the PS nanosphere. It is believed that the peak at about 31°

is strongly correlated with the whispering-gallery modes (WGMs) caused by the Mie resonance of individual nanospheres [33, 34]. Furthermore, the WGMs can propagate in the plane of the monolayer of spheres as a dielectric waveguide, as shown in Figure 5(d). Similar behavior has been reported in Ref. [35]. Therefore, the incident light with a certain angle beyond the critical angle can be coupled into the array of nanospheres to form the WGMs. The WGMs would subsequently be diffracted into the far field by the periodic structure as leaky modes. Thus, the natural characteristics of the nanosphere array provide an additional physical

FIGURE 5: The simulated relationships between transmission and incident angles at a wavelength of 385 nm for LEDs with and without a monolayer of PS nanospheres with a diameter of 400 nm under (a) s and (b) p polarizations. Simulations of the spatial distributions of electric field intensity for LEDs covered with a monolayer of PS nanosphere under p polarization at incident angles of (c) 23° and (d) 31°.

mechanism that increases light extraction. Additionally, several dips below the critical angle appeared in the transmission spectra. These dips resulted in some extra light being diffracted back into the LED. The back-diffracted light can be reflected by the back of the LED and then be re-extracted by the structures, which could raise the ultimate extraction efficiency [19].

The increase of the LEE of the NUV LEDs with a monolayer of hexagonal close-packed PS nanospheres is mainly caused by the following reasons. First, the periodic PS nanosphere arrays can act as 2D diffraction gratings to take out some guide modes that then propagate inside the LEDs through the near-field coupling. Second, when the size of the PS nanospheres is close to the emission wavelength, the guide modes can be coupled to the spheres with the evanescent field and diffracted into the far field by the periodic structure.

4. Conclusions

Monolayers of hexagonal close-packed PS nanospheres with different diameters were prepared on the ITO layer of NUV LEDs. The effect of PS nanospheres on the LEE of LEDs

was investigated. The results showed that a PS nanosphere monolayer on the transparent ITO electrode enhanced the NUV optical output of the LED. The maximum LEE was obtained when the nanosphere size was similar to the wavelength of light. Compared with that of the corresponding conventional LED without a PS nanosphere monolayer, the NUV LED with a monolayer of nanospheres of the optimum size resulted in an enhancement of LEE of 1.27-fold at $I = 100$ mA. The results of FDTD simulations and experiments agreed well. The calculated transmission spectrum showed that part of the incident light beyond the critical angle was extracted when the surface of NUV LEDs was covered with monolayer of PS nanospheres, leading to enhanced LEE. This approach using PS nanospheres has the advantages of low cost and high precision and is suitable for large-area preparation on LEDs. Thus, this is a simple and cost-effective approach to raise the efficiency of NUV LEDs.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that there is no conflict of interests.

Acknowledgments

We thank Prof. Zhang-Kai Zhou from Sun Yat-sen University for helpful discussions. This work was partly supported by the National Natural Science Foundation of China (Grant No. 61475038), Science and Technology Program of Guangzhou, China (Grant No. 201804010175), and Special Innovation Project of Department of Education of Guangdong Province (Grant No. 2018KTSCX123).

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