Enhanced light extraction efficiency via double nano-pattern arrays for high-efficiency deep UV LEDs

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ABSTRACT

This work reports on the packaging structure of double-layer nano-pattern arrays (NPAs) for AlGaN-based deep ultraviolet (DUV) light-emitting diodes (LEDs), which can significantly enhance the light extraction efficiency (LEE), a major device performance bottleneck. The double-layer NPAs were fabricated on the surface of the flip-chip DUV LED by etching sapphire and coating the fluoropolymer resin, which can alleviate the total internal reflection (TIR) at the top interfaces and enhance light extraction on sidewalls, leading to the great improvement of both TE and TM-polarized light. As a result, the 273 nm DUV LED with double-layer NPAs realized 28.3% enhancement in light output power (LOP) compared with the reference device without the NPA at large current of 100 mA. High peak external quantum efficiency (EQE) and wall-plug efficiency (WPE) of 5.19% and 4.38% were demonstrated, respectively. Combined with the finite element analysis (FEA), it is further confirmed that the double-layer NPAs have higher external coupling efficiency to enhance LEE. The proposed LEE enhancement strategy comes from the combination of nanostructured materials and packaging technology, which is cost-effective and meets mass production, providing efficient value in practical applications of UV devices.

1. Introduction

AlGaN-based deep ultraviolet (DUV) light-emitting diodes (LEDs) have attracted increasing attention due to potential applications of sterilization, medical phototherapy, secure communication, and so on [1,2]. However, the low light extraction efficiency (LEE) has limited the development of practical application for DUV LEDs. Firstly, the UV LEDs rely on AlGaN materials with high aluminum composition. Due to the spin-orbit effect and crystal field splitting, the topmost valence band is transformed from a heavy hole band to a crystal field splitting band as the Al component increases, leading to the predominant optical polarization of light emission is TM (E\textsubscript{\parallel}c) mode for DUV multiple quantum wells (MQWs) with high Al composition instead of TE (E\textsubscript{\perp}c) mode [3]. The propagation direction of TM-polarized DUV light is perpendicular to the c-axis, emitting mainly from the side of the MQWs. Therefore, the difficulty in extracting from the front side of the device limits the LEE of DUV LEDs [4]. Secondly, the excessive difference in refractive index exists in multiple nitride interfaces of flip-chip and sapphire/air interface, and the refractive index along the light emission path from MQWs to the air decreases successively. Therefore, a majority of DUV emission could be severely trapped inside the chip due to the existence of stronger reflection, especially total internal reflection (TIR), leading to the poor LEE [5]. Up to now, the typical value of LEE for DUV LEDs is still less than 10% [6], destining that there is still significant room for improvement. In comparison, the internal quantum efficiency (IQE) of DUV LEDs have reportedly exceeded 60% [7–10], implying that the main bottleneck limiting external quantum efficiency (EQE) and wall-plug efficiency (WPE) comes from the LEE. Therefore, improving the LEE is the most effective approach to enhancing the performance of DUV LEDs.

In order to optimize the light extraction of DUV LEDs, progress has been made by adopting multiple interfacial textures. Fujii et al promote a surface roughing on the n-side for GaN-based LED to improve the LEE, enhancing the surface emission successfully [11]. Gao et al introduce the highly reflective Cr/Au metal as the n-electrode to reflect the UV emission from the MQWs then improve the LEE [12]. Shkaya et al...
fabricate the photonic crystals on the mesa, which also could achieve the high reflectivity on the top interface and obtain the power improvement [13]. Xu et al combine the compressive in-plane strain and surface nanostructure that realize enhancement in facilitating TE mode emission [14]. However, the internal-level strategies also have potential problems such as high cost, sophisticated process, uniformity, etc., hindering further improvements and applications from implementing.

The packaging is an indispensable part of the fabrication process of DUV LEDs, but relatively less attention has been paid to it for exploring the potential of light extraction. In fact, the improvement of LEE could be realized by optimizing the packaging process to some extent [15,16]. In this paper, we combined superior polymer materials with nanoarray structures and proposed a packaging method by fabricating double-layer nano-pattern arrays (NPAs) to significantly enhance the LEE and polarization emission extraction of DUV LEDs via the modulation of interface optical field. Meantime, the theoretical finite element analysis (FEA) simulations were implemented to verify and analyze the physical mechanism. The advantages of this method include low cost, straightforward process, and effective LEE increase.

2. Experiment

2.1. Fabrication

Fig. 1 shows the fabrication step flow of the double-layer NPAs. The epitaxial wafers of DUV-LED were grown on sapphire substrate (2 in. in diameter) by metal-organic chemical vapor deposition (MOCVD) in advance, whose epitaxial structure is shown in Fig. 2(a). Then a deposited silicon dioxide layer on the non-epitaxial side of the sapphire substrate was used as a mask to improve the accuracy of pattern transfer. The positive photoresist was developed for 60 s to form nanoholes, and then the pattern is transferred to the SiO$_2$ film by inductively coupled plasma (ICP) etching with the mixture atmosphere of CF$_4$ and O$_2$ for 2 min, followed by the second ICP etching with the mixture atmosphere of Cl$_2$ and BCl$_3$ for 2 min. Afterwards, the first NPA layer was obtained followed by removing SiO$_2$ mask with Buffered Oxide Etch. Subsequently, the fluoropolymer resin with more than 95% transmittance at 273 nm was spined coating on the first NPA layer, followed by curing at 120 °C for 1 h and stripping to achieve the second NPA layer comprising hardened fluoropolymer resin. Moreover, the epitaxial wafers that have undergone standard chip fabrication processes were split into chips, which possess the size of about 8 × 20 mil for all devices. Finally, by flipping the second NPA film, cutting it to 3.5 × 2.5 mm, and coating it back on the first NPA and the sidewalls of chips (the last step), which applied additional fluoropolymer resin on the sidewall of the chip then curing to achieve adhesion, the DUV LEDs with double-layer NPAs was obtained. The flip-chips are fixed on a bracket, and then packaged on an AlN ceramic carrier by eutectic bonding [17] as shown in Fig. 2(b). Lately, the DUV LEDs devices without NPA, with single-layer NPA and double-layer NPAs are researched in this work for comparison.

2.2. Characterization and simulation

Field-emission scanning electron microscope (Nova Nano SEM FEI•F50) and atomic force microscope (AFM) were used to perform the morphology and profile depth of the NPA, indicating that the depth, period and radius of the NPA are about 350 nm, 1000 nm, 350 nm, respectively, as shown in Fig. 2(a). The electroluminescence (EL) spectra, the current-voltage, and light output power (LOP) of the LEDs were measured using an optoelectronic analysis system (ATA-1000, EVERFINE, Hangzhou, China) with a 30-cm-diameter integrating sphere at room temperature. Furthermore, the full spatial TE/TM mode light intensity was measured by a self-constructed test system with a Glan-Taylor prism, which placed between DUV-LED fixed on an angle-resolved bracket and spectrometer, as illustrated in Fig. 5. Furthermore, the FEA simulation was introduced to investigate the electromagnetic field distribution mode of DUV-LEDs without the NPA, with the single-layer NPA, and with the double-layer NPAs, which was
performed in an interactive 3D entity-modeling circumstance to shed light on the internal physical mechanism.

3. Results and discussion

The measurements of spectra, LOP and I-V curve of the DUV devices without the NPA, with the single-layer NPA and with the double-layer NPAs are conducted after encapsulated on the bracket. The devices emit at the same DUV peak wavelength of 273 nm shown in Fig. 3 since the NPA does not affect the emission wavelength. It is noting that the emission intensity increases with the coverage of NPA and rises to the maximum with the double-layer NPAs, providing preliminary verification of the enhancement of the NPA for light extraction. In Fig. 4(a), it could be seen clearly that the LOP of the devices reached saturation at 100 mA, which is 28.3% and 13.0% higher compared with the device without NPA and with the single-layer NPA, respectively. As a result, it should be noted that the devices almost possess the similar IQE [18]. Fig. 4(b) represents the devices also possess almost the same I-V characteristic and forward voltage (~5.4 V at 20 mA), which indicated that the NPA is not involved in the carrier transport process and confirmed the semblable current injection efficiency (CIE).

The EQE and WPE of the devices were calculated by Eqs. (1) and (2):
\[
\text{EQE} = \frac{\text{IQE} \cdot \text{LEE}}{I/V} = \frac{\text{LOP}}{h \nu}  \\
\text{WPE} = \frac{\text{EQE} \cdot \text{CIE}}{I/V} = \frac{\text{LOP}}{I/V}
\]

where \( h \) is Planck constant, and \( \nu \) is frequency of photon. From the calculation results illustrated in Fig. 4(c), both EQE and WPE of all devices obtained a maximum approximately at an injection current of 20 mA, exhibited an apparent enhancement in EQE and WPE of the double-layer NPAs compared with other devices, which rise up to 5.19% and 4.38%, respectively. Therefore, there are sufficient reasons to explain that the enhancement of WPE and EQE mainly lie in the effective improvement of LEE. Nevertheless, the efficiencies decrease with increasing injection current and the common phenomenon of efficiency
droop existed among nearly all III-nitride-based LEDs appears because of the current saturation. It is mainly attributed to excessively tiny scale of chips, the non-radiative Auger recombination caused by possible internal defects in epitaxial structure or the existence of electron leakage, lack of hole injection, and carrier delocalization [19,20]. Meanwhile, the LEE enhancement factor of the devices with single-layer NPA and double-layer NPAs under different drive currents as presented in Fig. 4 (d). The LEE enhancement factor from the NPA structures gradually increased as the current and emission intensity increases, and then maintained a high level at 50–70 mA, achieving a 35% enhancement benefit from the double-layer NPAs. However, it appears a downward trend after the current continues to increase. In this respect, the TE-polarized light radiated by the MQWs increases caused by increased heat accumulation in the chip [21], and then a part of the TE-polarized light propagating at a small angle along the c-axis direction might be strongly reflected by the NPA to a certain extent, reducing slightly the LEE enhancement factor.

To understand the far-field patterns of the DUV LEDs, the far-field emission intensity was angularly measured with a step size of 5°, performed by the light intensity test system (Fig. 5) without Glan-Taylor prism after the devices were fixed on the angle-resolved bracket. As shown in Fig. 6(a), the reference DUV LED has a Lambertian-like radiation patterns. As expected, it can be clearly seen that the emission envelopes of the device with single-layer NPA is greatly enhanced than that of the reference device without the NPA. The light propagation characteristics schematic for the different devices contribute to explain the enhancement principle, as shown in Fig. 6(b). The severe TIR exceeding 35.5° caused by the huge refractive index difference between sapphire ($n_s = 1.72$) and air ($n_a = 1$) appears on the interface without NPA, trapping the DUV emission inside. Nevertheless, the sapphire surface roughening by the NPA greatly enhanced light extraction from MQWs, which could be primarily attributed to the scattering by NPA on interface and the enlargement of emission angle [22]. Moreover, the enhancement of double-layer NPAs is more significant, revealing that NPA could effectively inhibit the TIR and enhance light extraction. For green and red curves, there are spikes at different angles, possibly due to photonic crystal (PC) effect caused by the NPAs.

Furthermore, the Glan-Taylor prism was added to the light intensity test system to measure spatial TE- and TM-polarized UV light separately

![Fig. 4.](image-url) (a) light output power, (b) I-V characteristic, (c) EQE and WPE, and (d) LEE enhancement factor of the DUV LEDs at different currents.

![Fig. 5.](image-url) The far-field emission distribution measurement system of the DUV LEDs and the principle of the polarized light analysis.
The principle of the polarized light spatial distribution measurement system is illustrated in Fig. 5, where the plane ACE is the chip plane and the plane ABD is the polarizer plane. The rotation angle $\theta$ of the angle-resolved bracket not only represents the angle of DUV LED plane and Glan-Taylor prism plane, but also corresponds to the degree of light propagation away from the c-axis. The light emitted by the DUV LED is collected by the optical fiber spectrometer through the polarizer ($\phi$ represents the rotation angle of the polarizer’s transmission axis), and then the spectra curve and LOP corresponding to each group ($\theta, \phi$) was obtained. Based on the spatial position relationship of $\theta$ and $\phi$, each measured intensity value can be decomposed in TE/TM mode. Therefore, the electric field mode oscillating along the transmissive axis AD is expressed in Eqs. (3) and (4):

\[
|E_{\text{TE}}| = |E_{\text{AE}}| = A_0 |E_{\text{AD}}| \sqrt{\cos^2 \phi \cos^2 \theta + \sin^2 \phi}
\]

\[
|E_{\text{TM}}| = |E_{\text{ED}}| = A_0 |E_{\text{AD}}| \cos \phi \sin \theta
\]

where $|E_{\text{AD}}| = A_0 \cdot LOP(\text{a.u.})$, and $A_0$ is a constant.

For TE polarization as presented in Fig. 7(a), the single-layer NPA device shows significantly stronger emission than the reference device because most of the UV light out of the non-epitaxial sapphire surface is TE-polarized whose electric field is perpendicular to the c-axis [23]. Hence, the maximum value of TE mode emission from MQWs was at $0^\circ$ and the intensity of TE polarization gradually decreased from $0^\circ$ to $90^\circ$. The sapphire surface roughening and enhancement of coupling probability [24–26] induced by the NPA greatly enhanced the TE mode light extraction from MQWs. More importantly, a dielectric layer with gradient refractive index could be formed at the interface of nanoarray structure [27]. Therefore, the emergent light passing through the interface of the NPA structure might be equivalent to entering a medium.
with a graded refractive index, which can effectively reduce Fresnel reflection or even TIR [28,29]. Furthermore, the enhancement is further boosted by the additional NPA layer, as evidenced by the strongest TE-polarized far-field emission of the double-layer NPAs device. The refractive index (n_i = 1.34) of the fluoropolymer resin layer inserted is between sapphire and air, further enlarging the extraction angle and area of bottom interface by suppressing TIR [30–32] as shown in Fig. 6 (b). Meanwhile, the top NPA achieves farther light extraction by suppressing reflection and increasing scattering again. For TM-polarized emission as presented in Fig. 7(b), it can hardly emit from the non-epitaxial sapphire surface due to its parallel electric field to the c-axis, whose intensity distribution is opposite to TE mode [23]. The multiple interfaces inside the epitaxial structure and sapphire/air interface can easily cause strong reflection of TM-polarized light, indicating that the enlarged extraction angle by NPA contributes rarely to the extraction of TM-polarized light. Thus, the far-field intensities of the reference device and the single-layer NPA device are similar. However, because the second NPA layer not only covers the top surface but also the sidewalls of the double-layer NPA device (Fig. 6(b)), it enhances the extraction of TM-polarized light which travels in-plane. As a result, the LEE improvement of the TM-polarized emission of the double-layer NPA LED is more profound. Essentially, the two refractive index gradients brought about by the opportune superposition of the two-layer high-frequency NPA structure effectively reduce the serious Fresnel reflection and TIR caused by the huge refractive index difference at the original sapphire/air interface, thereby achieving a significant improvement in light extraction of the double-layer NPAs.

The enhancement of both TE- and TM-polarized light is the most significant feature of the double-layer NPAs, resulting in strong overall far-field intensity in Fig. 6 and high device performance in Fig. 4. Besides, the far-field pattern of the DUV LED with single-layer NPA structure has strong disturbance, which is mainly caused by the PC structure [33]. On the contrary, the disturbance of the double-layer NPA structure is partially suppressed, which may be caused by imperfect alignment between the two NPA layers, resulting in much weaker PC scattering effect. Meanwhile, the simulation of multi-angle electromagnetic field distributions of the three devices was performed by FEA using the same devices and NPA dimensions. For the double-layer NPAs device, the air and resin cones are assumed to be aligned randomly, and then 273 nm light is incident on the three interfaces at 0°, 35.5° (TIR angle), 68° (large emergent angle) and the sidewall interface at 60° and 75°. As a result, the cross-sectional and the spatial electric field intensity distributions are shown in Fig. 8. It is noticed that the electric field distribution of the interface without NPA appears obvious feature of evanescent wave at or above 35.5°, implying the light trap inside caused by strong TIR. Conversely, the interface with NPA effectively breaks through the TIR trap and realizes the escape of the electric field vector into the air at the same emergent angle. Moreover, the addition of the second layer of NPA further lead to a more considerable electric field intensity in ambience, which also realized enhancement in flank by providing optical escape cones on the sidewall (Fig. 8(d)). It is demonstrated that the attachments of each layer NPA contribute to extra escaped lights benefit from additional refractive index gradient and light coupling. The spatial electric field distribution under the incident angle of 35.5° also verifies the entire enhancement process. Therefore, it is reasonable to believe that the double-layer NPAs have higher external coupling efficiency to enhance LEE, which is in general consistent with the experiments as discussed above.

4. Conclusion

In summary, we realize the 273 nm DUV LED chips with double-layer NPAs by applying nanoarray structures prepared by fluoropolymer resin amid the packaging process. In comparison to the reference flip-chip DUV LED, significant enhancement was obtained in the photo-electronic performance of DUV LED with double-layer NPAs which achieves maximum EQE and WPE of 5.19% and 4.38%, respectively. Furthermore, the DUV LED with the double-layer NPAs achieved an improvement of 28.3% in LOP than the reference LED without NPA. The underlying mechanism of LOP improvement of the DUV LEDs with different NPAs is analyzed by combing FEA simulation. It is attributed to the effective enlargement of extraction angle by reducing the TIR at top interfaces and enhancement by extraction cones on sidewall interface, leading to the external coupled emission improvement of both TE and TM polarization. It is believed that this packaging strategy combined with the preparation of novel nanoarray structural materials can provide neoteric research technique for LEE improvement and contribute to high-performance DUV LEDs in the future.

CRediT authorship contribution statement

Zhihua Zheng: Investigation, Methodology, Validation, Formal...

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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