

Growth of InGaN-based blue-LED on AlN/sapphire sputtered with different oxygen flow rate

Jiahui HU, Feng WU, Jiangnan DAI, Changqing CHEN (✉)

Wuhan National Laboratory for Optoelectronics, Huazhong University of Science and Technology, Wuhan 430074, China

© Higher Education Press 2021

Abstract Indium gallium nitride (InGaN) based blue light-emitting diodes (LEDs) suffer from insufficient crystal quality and serious efficiency droop in large forward current. In this paper, the InGaN-based blue LEDs are grown on sputtered aluminum nitride (AlN) films to improve the device light power and weaken the efficiency droop. The effects of oxygen flow rate on the sputtering of AlN films on sapphire and device performance of blue LEDs are studied in detail. The mechanism of external quantum efficiency improvement is related to the change of V-pits density in multiple quantum wells. The external quantum efficiency of 66% and 3-V operating voltage are measured at a 40-mA forward current of with the optimal oxygen flow rate of 4 SCCM.

Keywords light-emitting diode (LED), sputtered aluminum nitride (AlN), physical vapor deposition (PVD), metal-organic chemical vapor deposition (MOCVD)

1 Introduction

Due to the lack of native substrate, III-nitrides were grown on heteroepitaxial templates, such as sapphire, Si and SiC. The lattice mismatch of GaN and sapphire substrate is up to 16%, causing a high density of misfit dislocation. However, Ga atom tends to nucleate randomly on sapphire at the beginning of GaN deposition, resulting in the rough surface appearance of GaN films. In 1983, Yoshida et al. grew high-quality GaN films with an aluminum nitride (AlN) nucleation layer, which provides nucleation points for GaN deposition [1]. In 1991, Nakamura activated the p-type GaN by annealing the p-type GaN in a nitrogen atmosphere and fabricated GaN-based light-emitting diode (LED) for the first time [2]. In 1992, Nakamura et al.

improved the crystalline quality of GaN films with low-temperature GaN buffer [3]. In 1995, Nakamura et al. fabricated the InGaN-based LED with multiple quantum wells for the first time [4]. Then, the performance of InGaN-based LEDs was developed quickly with the industrialization process. Early in 2014, the electro-optical efficiency of InGaN-based LEDs was up to 303 lm/W [5].

Although great performance improvements have been made, InGaN-based LEDs suffer from serious efficiency droop in the huge forward current [6]. The easiest way to enhance the light power of LEDs is to increase the forward current, making more carriers transporting to multiple quantum wells; more carriers promote radiative recombination. However, when the forward current was increased to a certain value, which is different for LEDs, the light power of LEDs gradually saturated, meaning the luminous efficiency gradually decreased. This phenomenon is called efficiency droop. Therefore, most LED manufacturers must limit the working current of LEDs to avoid huge heat caused by the efficiency droop. The mechanisms of efficiency droop have been studied in detail [7–9]. On one hand, the electron overflow from multiple quantum wells to p-type layers becomes worse in huge forward current, which results in a decline of radiative recombination. On the other hand, the efficiency droop is proved to be related to Auger recombination in huge forward current.

To solve the above problems, many attempts have been made on InGaN-based LED structure design and GaN crystalline quality [10]. Tian et al. proposed an InGaN-based LED structure with gradually varied thickness of quantum barriers, which can improve the equivalent barrier height of electron blocking layer (EBL) [11]. Yen et al. optimized the thickness of quantum barriers to promote the carrier injection to multiple quantum wells [12]. Cheng and Wu replaced the general last barrier and EBL with a GaN/InGaN/GaN last barrier, which can impede electron overflow, improve hole injection and weaken efficiency droop effectively [13]. Besides device structure optimization, crystalline quality improvement is another way to

weaken the efficiency droop. An effective method is to fabricate the AlN buffer with physical vapor deposition (PVD). The PVD-AlN buffer is more uniform and compact than the conventional low-temperature AlN buffer deposited by metal-organic chemical vapor deposition (MOCVD), which is good for the crystalline quality of GaN films [14]. In this study, the effects of oxygen flow rate in the PVD-AlN buffer preparation on the crystalline quality of GaN films and the luminous efficiency of InGaN-based blue LEDs are studied in detail.

2 Experimental

During the deposition of AlN buffer in PVD, oxygen is needed to change the AlN film surface polarity. Beyond this, the film thickness and quality are affected by the oxygen flow rate. Hence, in this study, the effects of different oxygen flow rates are studied. Table 1 shows the oxygen flow rate used during the deposition of AlN buffer.

In this study, four samples called G501, G502, G503, and G504 are grown by MOCVD on PVD-AlN buffers with different oxygen flow rates. The oxygen flow rates for four samples are 2, 3, 4, and 5 SCCM. The device schematic is shown in Fig. 1. After the deposition of AlN buffer by PVD, 4.5- μm -thick n-type GaN was grown at 1050°C and 150 Torr. The electron concentration of n-GaN is $5 \times 10^{18} \text{ cm}^{-3}$. Then, eight pairs of GaN/InGaN multiple quantum wells were grown on the n-GaN template at

Table 1 Oxygen flow rate during AlN buffer deposition for different samples

sample	device power/kW	gas	oxygen flow rate/SCCM
G501	5	N ₂ , Ar, O ₂	2
G502	5	N ₂ , Ar, O ₂	3
G503	5	N ₂ , Ar, O ₂	4
G504	5	N ₂ , Ar, O ₂	5

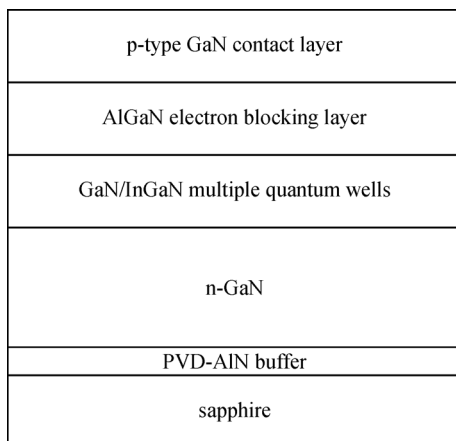


Fig. 1 Device schematic of LED on PVD-AlN buffer

860°C and 200 Torr. The content and thickness of the InGaIn quantum well are 15% and 3 nm, respectively. The thickness of the GaN quantum barrier is 11 nm. The quantum well and barrier are intrinsic. Following the multiple quantum wells is a 30-nm thick p-type AlGaIn electron blocking layer at 915°C and 100 Torr. The Al content of EBL is 15%, and the hole concentration is $1 \times 10^{20} \text{ cm}^{-3}$. The top layer of LED is a 50-nm-thick p-type GaN contact layer, and the hole concentration is $1 \times 10^{19} \text{ cm}^{-3}$. The growth temperature and chamber pressure of p-type GaN are 950°C and 500 Torr, respectively. After the growth of the wafer, 380 $\mu\text{m} \times 760 \mu\text{m}$ LED chips were prepared by the standard chip process.

3 Results and discussion

The crystalline quality of all samples is measured by high-resolution X-ray powder diffraction (XRD), and the results are shown in Table 2. The (002) full width at half maximum (FWHM) of XRD for four samples is gradually increased, whereas the (102) FWHM is gradually decreased. The (002) FWHM of XRD for four AlN buffers is also shown in Table 2. To shed light on the change in crystalline quality of four AlN buffers, transmission electron microscopy (TEM) photos of four samples are shown in Fig. 2. The TEM photos illustrate that, the increased oxygen flow rate makes PVD-AlN buffer thinner. The increased thickness of PVD-AlN buffer promotes crystalline quality improvement, as shown in the XRD results. The XRD data demonstrates that the crystalline quality of GaN templates is gradually improved with an increase in oxygen flow rate from 2 to 5 SCCM. Generally, the thinner the PVD-AlN buffer thickness, the thicker was the GaN coalescence layer at the beginning of growth. Further coalescence can promote dislocation annihilation during the growth of the GaN template. The thickness of PVD-AlN buffer is gradually reduced with an increase of oxygen flow rate, improving crystalline quality. The stress state of four samples is measured using XRD (105) reciprocal space mapping (RSM). Figure 3 illustrates that the multiple quantum wells of all samples are completely strained.

To exhibit the interface between the PVD-AlN buffer and the GaN template, energy dispersive spectrometry (EDS) mapping was conducted near the PVD-AlN buffer,

Table 2 XRD rocking curve FWHM of samples G501, G502, G503, and G504

sample	AlN(002)/arcsec	GaN(002)/arcsec	GaN(102)/arcsec
G501	9.9	119.5	185.9
G502	10.2	127.9	175.5
G503	16.3	128.3	173.8
G504	22.8	130.6	166.5

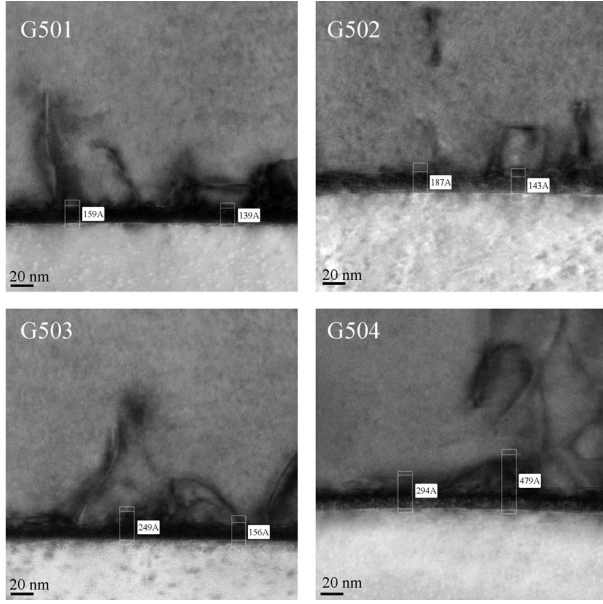


Fig. 2 TEM results of samples G501, G502, G503, and G504

as shown in Fig. 4. The area between the yellow dotted line is the PVD-AlN buffer. We demonstrated that the interface between the AlN and GaN is clear. No O is in the PVD-

AlN buffer, meaning that oxygen is just a way to provide an oxygen-enriched environment, not a source. The surface morphologies of the four samples are measured using atomic force microscopy (AFM), as shown in Fig. 5. The root mean square (RMS) of G501, G502, G503, and G504 is 0.368, 0.232, 0.185, and 0.158 nm, respectively.

The external quantum efficiency (EQE) and light output power of all packaged chips are measured by integrating the sphere test system, which has been averaged and shown in Fig. 6(a). We demonstrated that the EQE of sample G502 is highest in the four samples, which is 66%, whereas the lowest EQE is 62% for sample G501. The EQEs of G502 and G503 is nearly the same, whereas the EQE of G504 is lower than that of G502 and G503. However, the crystalline quality of G504 is better than that of G502 and G503 from the XRD measurement in Table 2, meaning that the crystalline quality improvement is not the key point for the increase in EQE. In addition, the efficiency droop for the four samples was 5.5%, 2.8%, 2.6%, and 3.1%, which is defined as the difference value between the max EQE and EQE at the 50-mA forward current. The optimization of oxygen flow rate in the preparation of PVD-AlN buffer is related to the efficiency droop is shown. In Fig. 6(b), the light power at different current is shown for the four samples. For sample G503,

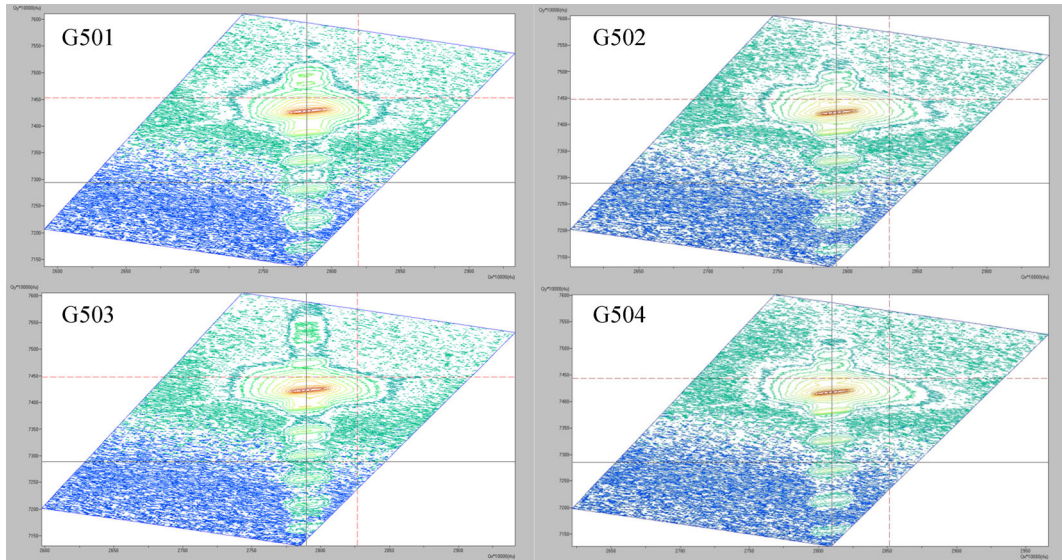


Fig. 3 XRD (105) reciprocal space mapping of samples G501, G502, G503, and G504

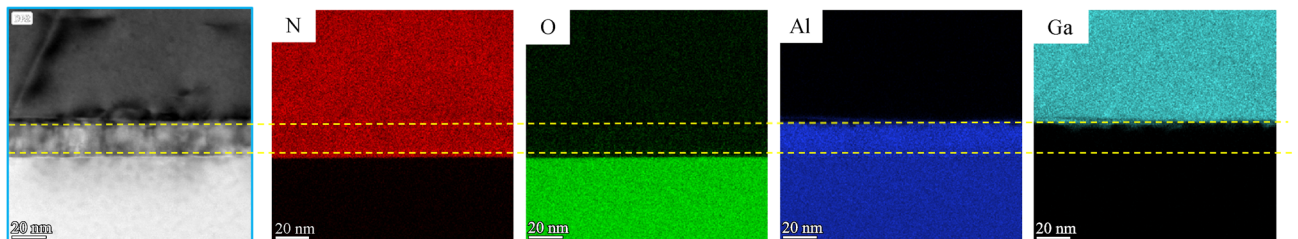


Fig. 4 EDS mapping for the sample G504

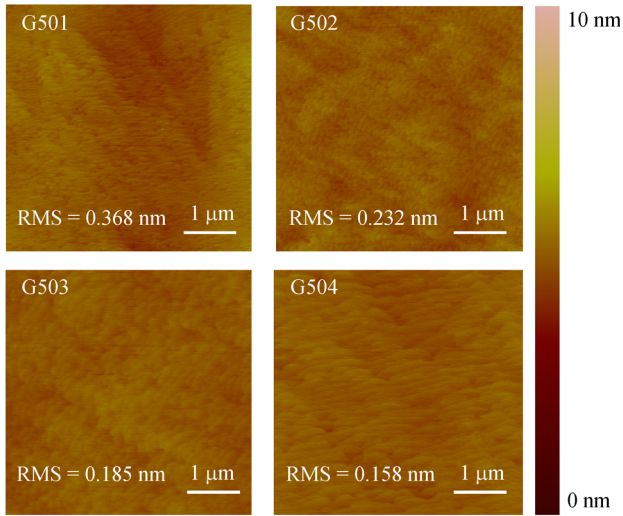


Fig. 5 AFM measurement for G501, G502, G503, and G504

the light power is 46.87 mW at 40 mA. The current–voltage (I – V) characteristics for four samples are shown in Fig. 6(c). At the 40-mA forward current, the voltages for

the four samples are gradually decreased from 3.2 V for G501 to 3 V for G504. The electroluminescence (EL) spectra of the four samples are shown in Fig. 6(d).

To shed light on the improvement of EQE, cathodoluminescence (CL) spectroscopy for four samples was measured and the results are shown in Fig. 7. It is exhibited that the densities of surface defects of the four samples are 2.75×10^8 , 1.65×10^8 , 1.59×10^8 , and $1.09 \times 10^8 \text{ cm}^{-2}$, respectively. Generally, the surface defects in the CL test are considered as the V-pits in blue LED, which play a vital role in the device performance. The morphology of V-pits is measured using TEM, as shown in Fig. 8. V-pits are like an aperture near the multiple quantum wells, forming an enrichment area, which can promote hole injection to the quantum wells near the n-type layer and improve the luminous efficiency. The luminous efficiency will improve with an increase in the density of V-pits. Alternatively, V-pits will result in the loss of light areas, causing the light power to decrease [15–20]. According to the density of V-pits, assuming the V-pit is approximately 200-nm long, the losses of light areas for G501, G502, G503, and G504 are 8.5%, 5%, 5%, and 3%. Hence, the injection promotion and light area loss both

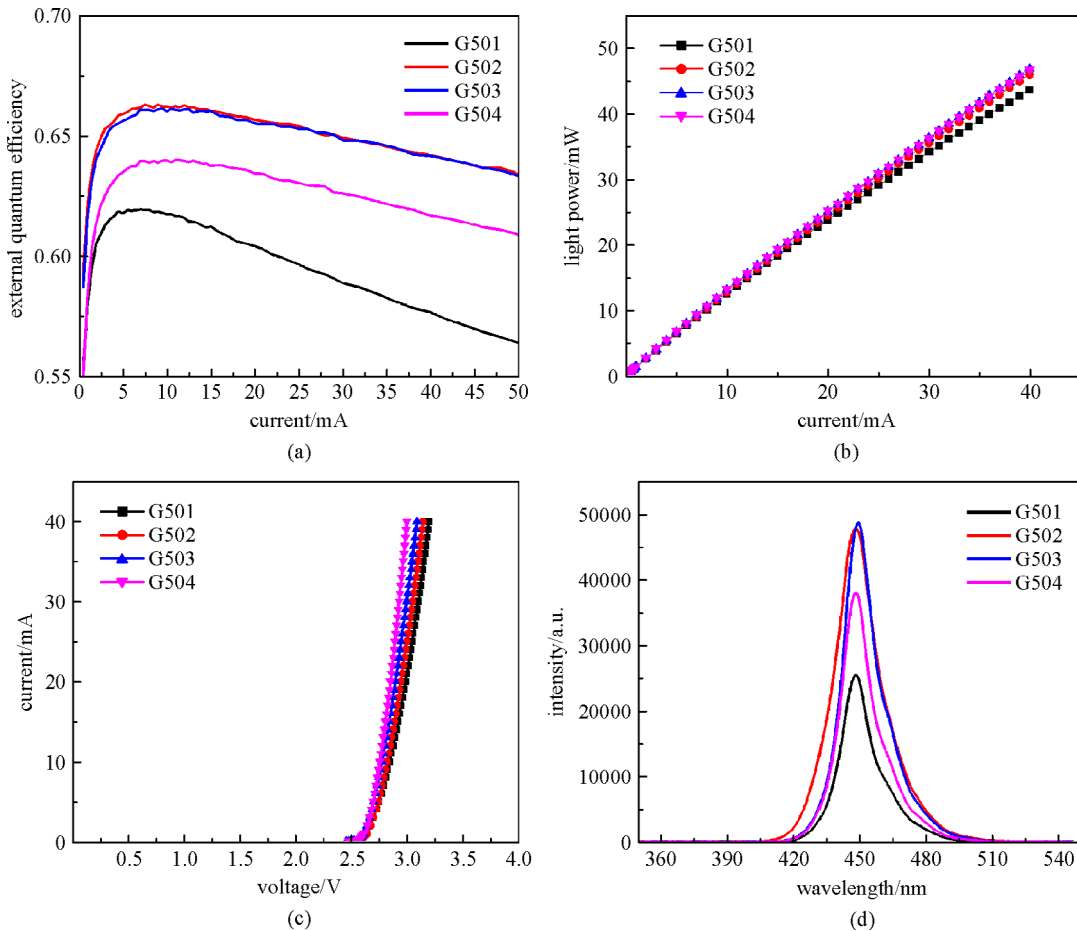


Fig. 6 (a) External quantum efficiency (EQE), (b) light power, (c) I – V characteristics, and (d) EL spectra for samples G501, G502, G503, and G504

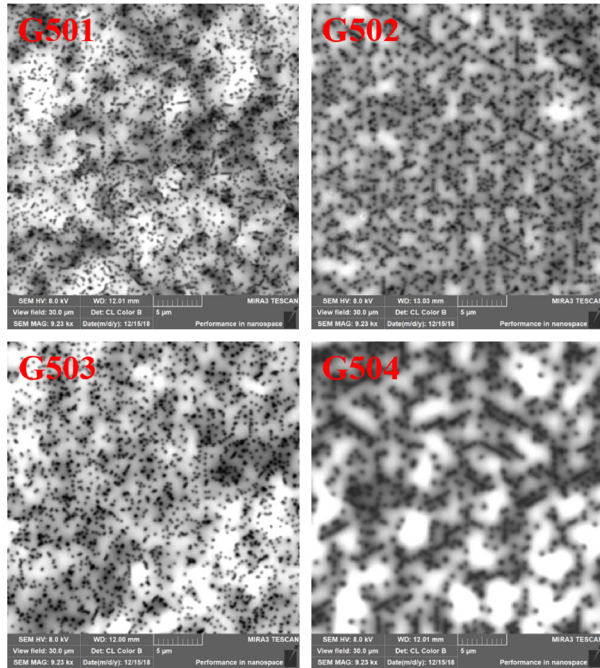


Fig. 7 Cathodoluminescence spectroscopy for samples G501, G502, G503, and G504

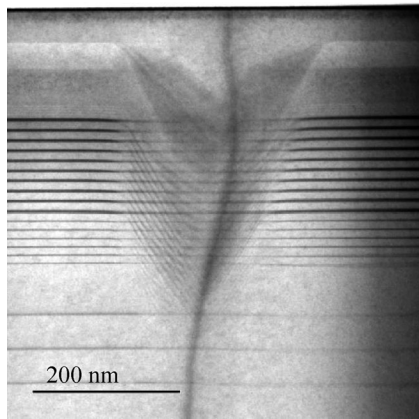


Fig. 8 TEM test for the surface defects in cathodoluminescence spectroscopy

give rise to the highest EQE for G503. In addition, the light area loss may cause an increase in actual current ampere density, which will further aggravate the efficiency droop. In conclusion, the oxygen flow rate of PVD-AlN buffer deposition could change the density of V-pits, which will further influence the luminous efficiency and efficiency droop.

4 Conclusions

The effects of oxygen flow rate on the sputtering of AlN films on sapphire and the device performance of blue

LEDs are studied in detail. The mechanism of EQE improvement is related to the change of V-pits density in multiple quantum wells, which is related to the hole injection promotion and light area loss. The EQE of 66% and operating voltage of 3 V are measured at a forward current of 40 mA with the optimal oxygen flow rate of 4 SCCM.

Acknowledgements The work was supported by the National Key Research and Development Program of China (No. 2019YFA0708203), the Center of Micro-Fabrication and Characterization (CMFC) of WNLO, Chinese Academy of Sciences (No. IIMDKFJJ-17-09), the National Natural Science Foundation of China (Grant Nos. 61704062, 61774065, and 61704176), and the China Postdoctoral Science Foundation (No. 2016M602287).

References

1. Yoshida S, Misawa S, Gonda S. Improvements on the electrical and luminescent properties of reactive molecular beam epitaxially grown GaN films by using AlN-coated sapphire substrates. *Applied Physics Letters*, 1983, 42(5): 427–429
2. Nakamura S. GaN growth using GaN buffer layer. *Japanese Journal of Applied Physics*, 1991, 30(Part 2, No. 10A): L1705–L1707
3. Nakamura S, Mukai T, Senoh M, Iwasa N. Thermal annealing effects on p-type Mg-doped GaN films. *Japanese Journal of Applied Physics*, 1992, 31(Part 2, No. 2B): L139–L142
4. Nakamura S, Mukai T, Senoh M. High-brightness InGaN/AlGaIn double-heterostructure blue-green-light-emitting diodes. *Journal of Applied Physics*, 1995, 76(12): 8189–8191
5. Amano H. Growth of GaN on sapphire via low-temperature deposited buffer layer and realization of p-type GaN by Mg doping followed by low-energy electron beam irradiation. *International Journal of Modern Physics B*, 2015, 29(32): 1530015
6. Shen Y C, Mueller G O, Watanabe S, Gardner N F, Munkholm A, Krames M R. Auger recombination in InGaIn measured by photoluminescence. *Applied Physics Letters*, 2007, 91(14): 141101
7. Iveland J, Martinelli L, Peretti J, Speck J S, Weisbuch C. Direct measurement of Auger electrons emitted from a semiconductor light-emitting diode under electrical injection: identification of the dominant mechanism for efficiency droop. *Physical Review Letters*, 2013, 110(17): 177406
8. Kim M H, Schubert M F, Dai Q, Kim J K, Schubert E F, Piprek J, Park Y. Origin of efficiency droop in GaN-based light-emitting diodes. *Applied Physics Letters*, 2007, 91(18): 183507
9. Piprek J. Efficiency droop in nitride-based light-emitting diodes. *Physica Status Solidi A, Applications and Materials Science*, 2010, 207(10): 2217–2225
10. Wang J, Chen Z, Xing Y, Wang L, Luo Y. The influences of sputtered AlN buffer layer on AlInGaIn based blue and near-ultraviolet light emitting diodes. *Physica Status Solidi A, Applications and Materials Science*, 2017, 214(6): 1600714
11. Tian W, Zhang J, Wang Z, Wu F, Li Y, Chen S C, Xu J, Dai J N, Fang Y Y, Wu Z H, Chen C Q. Efficiency improvement using thickness-chirped barriers in blue InGaIn multiple quantum wells light emitting diodes. *IEEE Photonics Journal*, 2013, 5(6): 8200609

12. Yen S H, Tsai M L, Tsai M C, Chang S J, Kuo Y K. Investigation of optical performance of InGaN MQW LED with thin last barrier. *IEEE Photonics Technology Letters*, 2010, 22(24): 1787–1789
13. Cheng L, Wu S. Performance enhancement of blue InGaN light-emitting diodes with a GaN-AlGaIn-GaN last barrier and without an AlGaIn electron blocking layer. *IEEE Journal of Quantum Electronics*, 2014, 50(4): 261–266
14. Valcheva E, Paskova T, Tungasmita S, Persson P, Birch J, Svedberg E B, Hultman L, Monemar B. Interface structure of hydride vapor phase epitaxial GaN grown with high-temperature reactively sputtered AlN buffer. *Applied Physics Letters*, 2000, 76(14): 1860–1862
15. Xu Y, Wu Q F, Zhou S J, Pan S, Wu X M, Zhang J L, Quan Z J. Influence of V-shaped pits on hole current distribution in GaN-based green LED. *Chinese Journal of Luminescence*, 2018, 39(5): 674–680
16. Koike K, Lee S, Cho S R, Park J, Lee H, Ha J S, Hong S K, Lee H J, Cho M W, Yao T. Improvement of light extraction efficiency and reduction of leakage current in GaN-based LED via V-pit formation. *IEEE Photonics Technology Letters*, 2012, 24(6): 449–451
17. Xu C, Zheng C, Wu X, Pan S, Jiang X, Liu J, Jiang F. Effects of V-pits covering layer position on the optoelectronic performance of InGaIn green LEDs. *Journal of Semiconductors*, 2019, 40(5): 61–65
18. Cho J, Schubert E F, Kim J K. Efficiency droop in light-emitting diodes: challenges and countermeasures. *Laser & Photonics Reviews*, 2013, 7(3): 408–421
19. Zhao Y, Fu H, Wang G T, Nakamura S. Toward ultimate efficiency: progress and prospects on planar and 3D nanostructured nonpolar and semipolar InGaIn light-emitting diodes. *Advances in Optics and Photonics*, 2018, 10(1): 246–308
20. Weisbuch C, Piccardo M, Martinelli L, Iveland J, Peretti J, Speck J S. The efficiency challenge of nitride light-emitting diodes for lighting. *Physica Status Solidi A, Applications and Materials Science*, 2015, 212(5): 899–913



Jiahui Hu received a Bachelor's degree from Xi'an Jiaotong University, China. He is currently pursuing a Ph.D. degree with the Wuhan National Laboratory for Optoelectronics, Huazhong University of Science and Technology, China. His research interests focus on the material growth and device structure design of blue and deep ultraviolet LEDs.



Dr. **Feng Wu** received his B.Sc. and Ph.D. degree in Optics Engineering from Huazhong University of Science and Technology, China, in 2012 and 2017, respectively. He then joined Prof. Weida Hu's group as a post-doctor in Shanghai Institute of Technical Physics, Chinese Academy of Sciences, China. Currently, he is an associate professor at Huazhong University of Science and Technology. His research interests focus on the growth and characterization of wide bandgap semiconductors and two-dimensional materials, and the corresponding optoelectronic devices such as LED, laser, and photodetectors. He has authored and co-authored more than 40 technical journal papers and conference presentations.



Prof. **Jiangnan Dai** received his Ph.D. degree from Nanchang University, China. He is currently a professor at Wuhan National Laboratory for Optoelectronics, Huazhong University of Science and Technology, China. His research interests are the wide gap band III-V group-based semiconductor materials and devices. He has devoted his research efforts to material growth and device preparation of semiconductor light-emitting diode (LED) and semiconductor laser diode (LD).



Prof. **Changqing Chen** received his B.Sc. degree in Applied Physics, M.Sc. degree in Semiconductor Physics and Devices from Wuhan University, China in 1992 and 1994, respectively. As a scholarship holder of DAAD Volkswagen Foundation, he achieved his Ph.D. degree in Semiconductor Physics and Devices from the University of Erlangen-Nürnberg, Germany in 2000. He joined Huazhong University of Science and Technology and accepted the professorship in 2007. He has published more than 100 peer-reviewed SCI papers. He has devoted his research efforts to III-nitride materials and devices including ultraviolet LEDs and ultraviolet photodetectors.