

Recent progresses on InGaN quantum dot light-emitting diodes

Lai WANG (✉), Wenbin LV, Zhibiao HAO, Yi LUO

Tsinghua National Laboratory for Information Science and Technology, Department of Electronic Engineering,
Tsinghua University, Beijing 100084, China

© Higher Education Press and Springer-Verlag Berlin Heidelberg 2014

Abstract InGaN quantum dots (QDs) have attracted many research interests in recent years for their potentials to realize long wavelength visible emission from green to red, which can pave a way to fabricate the phosphor-free white light emitting diodes (LEDs). In this paper, we reported our recent progresses on InGaN QD LEDs, the discussions were dedicated to the basic physics model of the strain relaxation in self-assembled InGaN QDs, the growth of InGaN QDs with a growth interruption method by metal organic vapor phase epitaxy, the optimization of GaN barrier growth in multilayer InGaN QDs, the demonstration of green, yellow-green and red InGaN QD LEDs, and future challenges.

Keywords quantum dot (QD), InGaN, light emitting diode (LED), quantum confined Stark effect (QCSE)

1 Introduction

InGaN materials have very important applications in the fields such as energy saving and environmental protection [1]. It is current one of the research focuses of semiconductor optoelectronic materials. InGaN quantum dots (QDs) behave nanoscale in three dimensional directions, resulting in quantum confinement of the three directions. Compared to quantum well (QW) structure, QDs exhibit more excellent properties in some areas, such as light emitting diodes (LEDs) [2] and single photon source photoelectric devices [3]. Therefore, InGaN QDs have been widely investigated in basic physics and application devices, for their unique properties.

In this paper, we focused on the applications of InGaN QD LEDs. As is well known, “green gap” is a troublesome problem in the research area of LEDs [4]. Many

publications reported the measures of overcoming “green gap” by changing the InGaN growth plane of QW structure from the c-plane to nonpolar [4–6] or semipolar planes [7–10]. Although great progress has been made in the nonpolar and semipolar LEDs, there are still some shortcomings. Nonpolar and semipolar growth of InGaN on sapphire substrate made higher density of dislocations and stacking faults than traditional c-plane [11,12]. On the other hand, in order to improve the crystal quality, expensive nonpolar and semipolar GaN substrates are desired [13], but it is still difficult to obtain large size of nonpolar and semipolar GaN substrates. Therefore, the nonpolar and semipolar LEDs cost significantly higher than c-plane. In addition, although the polarization field can be ignored in nonpolar or semipolar InGaN/GaN multiple QWs, considering the large lattice mismatch between InGaN and GaN, this will lead to much strain, especially when InGaN QWs are wide or composition of indium is high.

In fact, strain can be weakened by InGaN QDs, resulting in the reduction of quantum confined Stark effect (QCSE). Compared to the nonpolar or semipolar plane solutions, c-plane InGaN QDs growth does not cause additional stacking fault. More importantly, it is compatible to the InGaN based conventional growth process with low cost manufacturing factory.

2 Theoretical study on critical thicknesses of InGaN grown on (0001) GaN

As InGaN grown on the GaN, there is a huge stress caused by the lattice mismatch, and stress release mechanism for epitaxial growth is very important. For epitaxial growth of InGaN, there are two kinds of stress release mechanism [14–16]: one is through the dislocation to release stress [15]; the other is through the surface morphology changes from two-dimensional to three-dimensional [16]. Two

kinds of stress release mechanism correspond to different critical thicknesses. Based on the physical model between the strain energy and the thickness of InGaN, the critical thicknesses with different indium composition are shown in Fig. 1 in the two mechanisms [14].

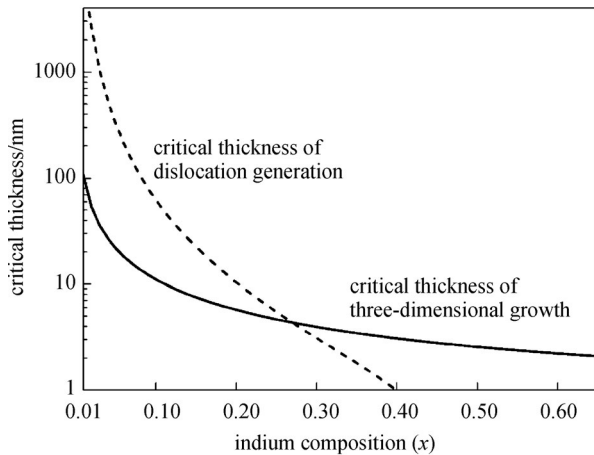


Fig. 1 Critical thicknesses of dislocation generation and three-dimensional growth versus indium composition x ($\text{In}_x\text{Ga}_{1-x}\text{N}$) while InGaN is grown on (0001) GaN. After W. Zhao et al. [14]. Reprinted from Journal of Crystal Growth, 327, W. Zhao et al., pp. 202. Copyright (2011), with permission from Elsevier

When the indium composition is less than 27%, the critical thickness of the three-dimensional growth is less than that of the dislocations. When the indium composition is relatively low, InGaN stress is released preferentially through the three-dimensional growth without generating dislocations. On the other hand, when indium composition is more than 27% in the generated dislocations model, the critical thickness decreases rapidly compared to three-dimensional growth model. So the stress will be released preferentially by dislocations. In the long-wavelength emitting ranging from green to red, the indium composition of the InGaN QDs is usually greater than 30%. It is easy to produce dislocations, but not conducive to the three-dimensional growth in these conditions.

3 Growth interruption method

To achieve InGaN QDs growth rather than dislocations, the growth interruption method [17–26] is used in the high indium composition of InGaN QDs. It includes three steps as shown in Fig. 2. First, a very thin layer of InGaN film is grown first on the surface of GaN, while the stress has not yet been released. Second, the growth process is interrupted. During the growth interruption, the InGaN layer is annealed and some parts of the thin InGaN layer decomposed. Some parts of indium and gallium atoms migrate in the surface, forming nanoscale islands to release

the stress. The process of interruption is the key point of the formation of the InGaN QDs. Third, another thin InGaN layer deposited, which can adjust the size and density of QDs. If the formation of QDs morphology meets the requirements in the second step, the third step can no longer be needed.

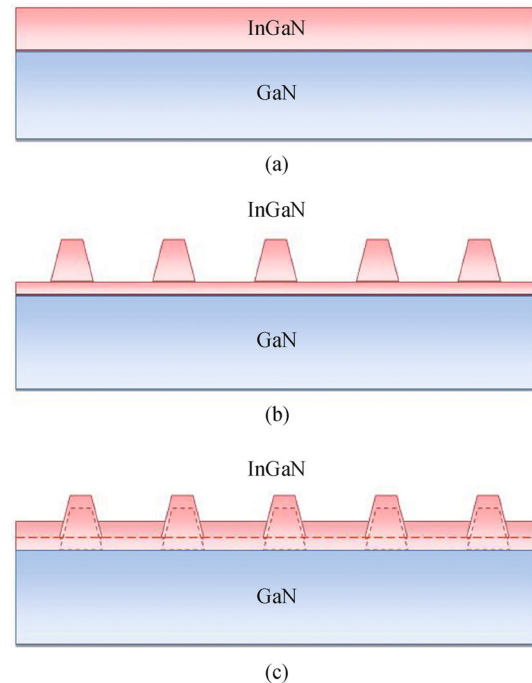


Fig. 2 Schematic diagram of the whole growth interruption process. The growth interruption method may include three steps: (a) deposition of InGaN film; (b) interruption of growth and annealing of InGaN to form dots; (c) continuation of growth of InGaN dots. After W. B. Lv et al. [26]. Reprinted from Japanese Journal of Applied Physics, 52, W. B. Lv, et al., pp. 08JG13. Copyright (2013), with permission from The Japan Society of Applied Physics

4 Self-assembled growth of InGaN QDs

Grown by the growth interruption method, the typical high indium composition InGaN QDs are shown in Fig. 3(a), in which the diameter, height and density are 14.3 nm, 7.6 nm, and $8 \times 10^9 \text{ cm}^{-2}$, respectively. Photoluminescence (PL) spectra under different temperatures are shown in Fig. 3(b). The 490 nm luminescence peak corresponds to InGaN QDs, and the low temperature 400–440 nm PL range corresponds to the InGaN wetting layer, which means the growth pattern is Stranski–Krastanov (S-K) growth mode. Figure 3(c) shows that the QD PL normalized integrated intensities increases first and then decreases while increasing temperatures, the full width at half maximum (FWHM) decreases first and then increases, which accords with the typical luminescence characteristics of S-K growth mode QDs. In addition, the sharp emission spectrum is clearly indicated in the low

temperature micro-PL, which fully proved the existence of QDs, as shown in Fig. 3(d).

5 Optimization of GaN barrier growth in multilayers QDs

The difficult of multilayer InGaN QDs epitaxial growth is that GaN barrier layers should be grown into smooth surface on the InGaN rough surface, so that the subsequent QDs layer can be grown on the same surface state. If the growth parameters of GaN barrier layer are the same as those of InGaN QDs, GaN surface are very rough (surface roughness of 1.937 nm), as shown in Fig. 4(a). Based on the rough GaN surface, multilayer QDs become very uneven, as shown in cross-sectional scanning transmission electron microscopy (TEM) dual layer QDs of Fig. 4(b).

Therefore, the GaN barrier layer growth parameters should be optimized. By changing the growth of carrier gas

from nitrogen to hydrogen and increasing the growth temperature of GaN barrier, the surface morphology of GaN barrier layer (GaN flat surface roughness of 0.416 nm), are shown in Fig. 5(a). Hence, ten layers of InGaN QDs were grown by the optimized growth parameters. The cross-sectional TEM image of ten layers InGaN QDs are shown in Fig. 5(b). The ten layers of uniform QDs and the flat surface of GaN barrier layer can be observed clearly. The diameter, height and density of every layer of QDs maintain the same level.

6 InGaN QD LEDs

In the range from green to red emission, high quality of high indium composition InGaN thin film is very difficult to grow. Due to the stress effect, piezoelectric polarization induced QCSE has become very serious. Using QDs instead of QWs can release partial stress, which may be

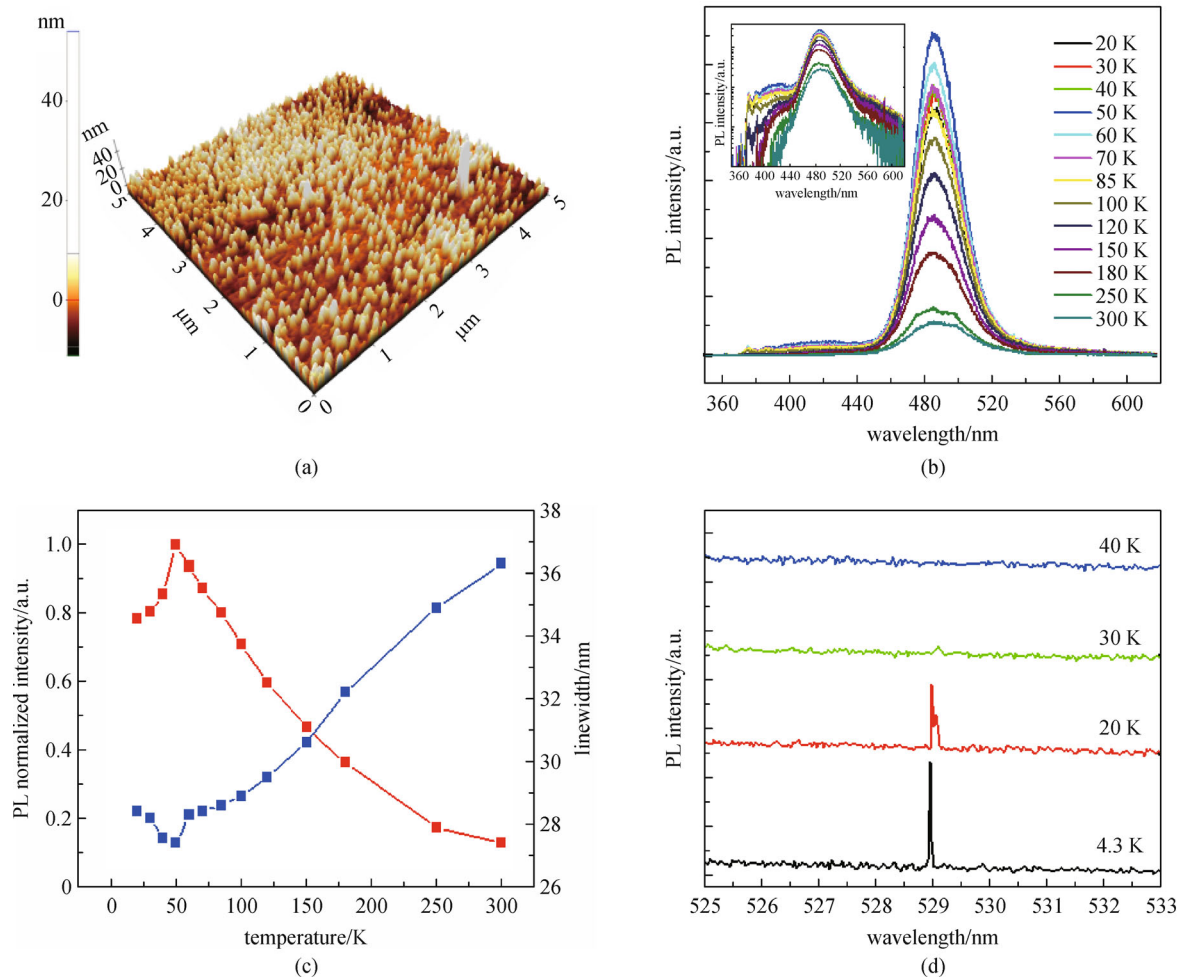


Fig. 3 (a) Atomic force microscopy (AFM) image of InGaN QDs; (b) temperature-dependent PL spectra, the inset in (b) are plot of logarithmic vertical axis scale; (c) normalized intensity and FWHM under different temperatures; (d) micro-PL spectra of a single QD at low temperature. After L. Wang et al. [27]. Reprinted from *Physica Status Solidi C*, 9, L. Wang, et al., pp. 782. Copyright (2012), with permission from Wiley

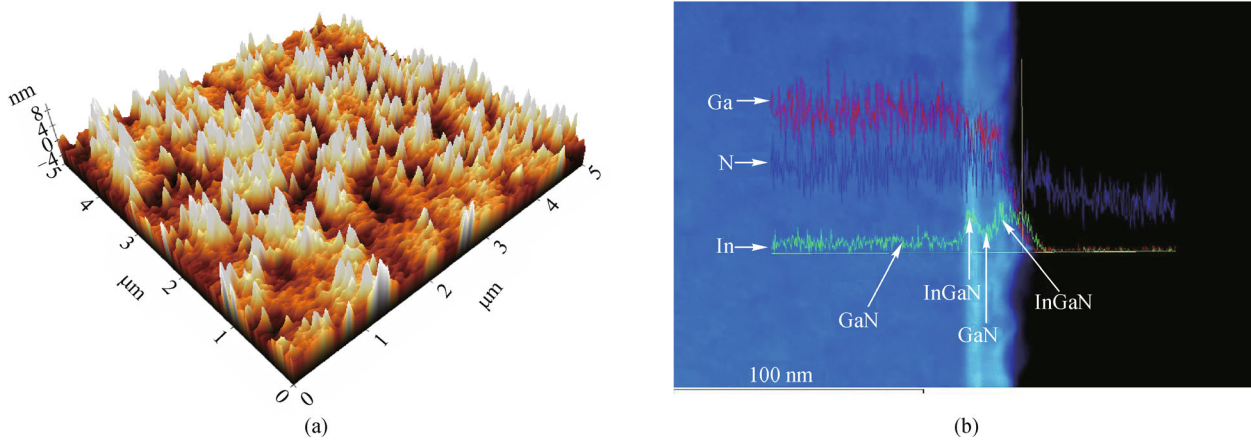


Fig. 4 (a) AFM image of rough GaN surface with unoptimized parameters; (b) Z-contrast scanning TEM image of dual layer InGaN QDs with unoptimized parameters. After W. B. Lv et al. [28]

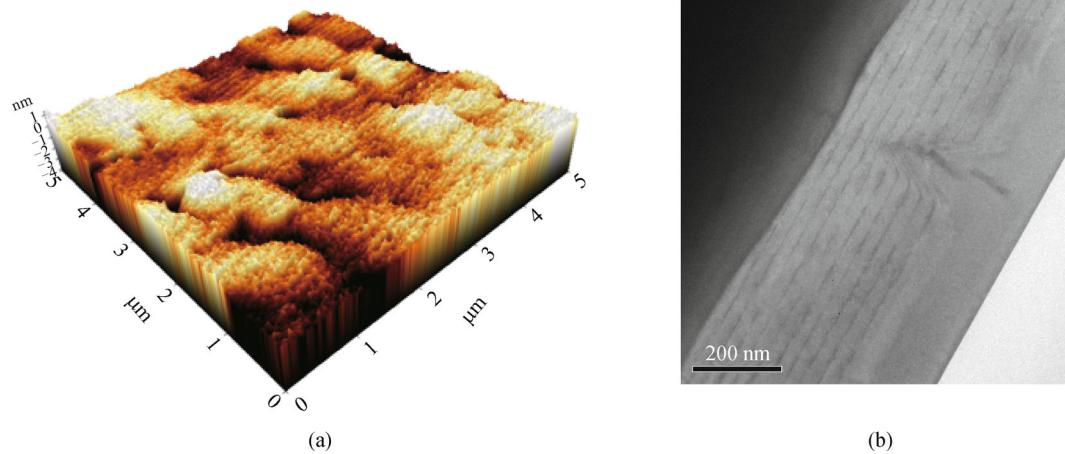


Fig. 5 (a) AFM image of flat GaN surface with optimized parameters; (b) cross-section TEM image of ten layers of InGaN QDs. After W. B. Lv et al. [29]

used to prepare long wavelength of visible light emitting devices with reduced QCSE. Depending on the growth interruption method and optimized GaN barrier growth situation, green, yellow-green and red LEDs are successfully grown by metalorganic vapor phase epitaxy (MOVPE) respectively. From Fig. 6, the electroluminescence (EL) peak wavelength is 527 nm at 20 mA for green LED, 543 nm at 20 mA for yellow-green LED and 729 nm at 80 mA for red LED, respectively. For green LED, the shift of peak wavelength is only 1.5 nm. The quite small blue shift means that the QCSE in the QDs is well suppressed [30]. For yellow-green LED, the amount of blue shift is comparable to that of yellow-green LEDs grown on semipolar planes [29]. For deep-red LED, the long wavelength emission is attributed to the composition pulling effect by the two factors including the InGaN QD structure and beneath InGaN/GaN super lattice [26]. It indicates a potential method of implementing phosphor free white LEDs in InGaN material system.

7 Future challenges

7.1 Dislocations in QDs

In fact, there are many dislocations in the interface between the GaN barriers and high indium composition InGaN QDs. In the first step of the growth process, the stress relieves through dislocations before forming InGaN QDs in the growth interruption. So it is beneficial to highly achieve indium incorporation in the growth interruption method. But the relationship between dislocations and QDs still needs further investigation. Whether the dislocations result in the poor performance of high indium InGaN QD devices, it has not been confirmed in the present study.

7.2 Performance of InGaN QD LEDs

As for the green InGaN QD LEDs, the light output power

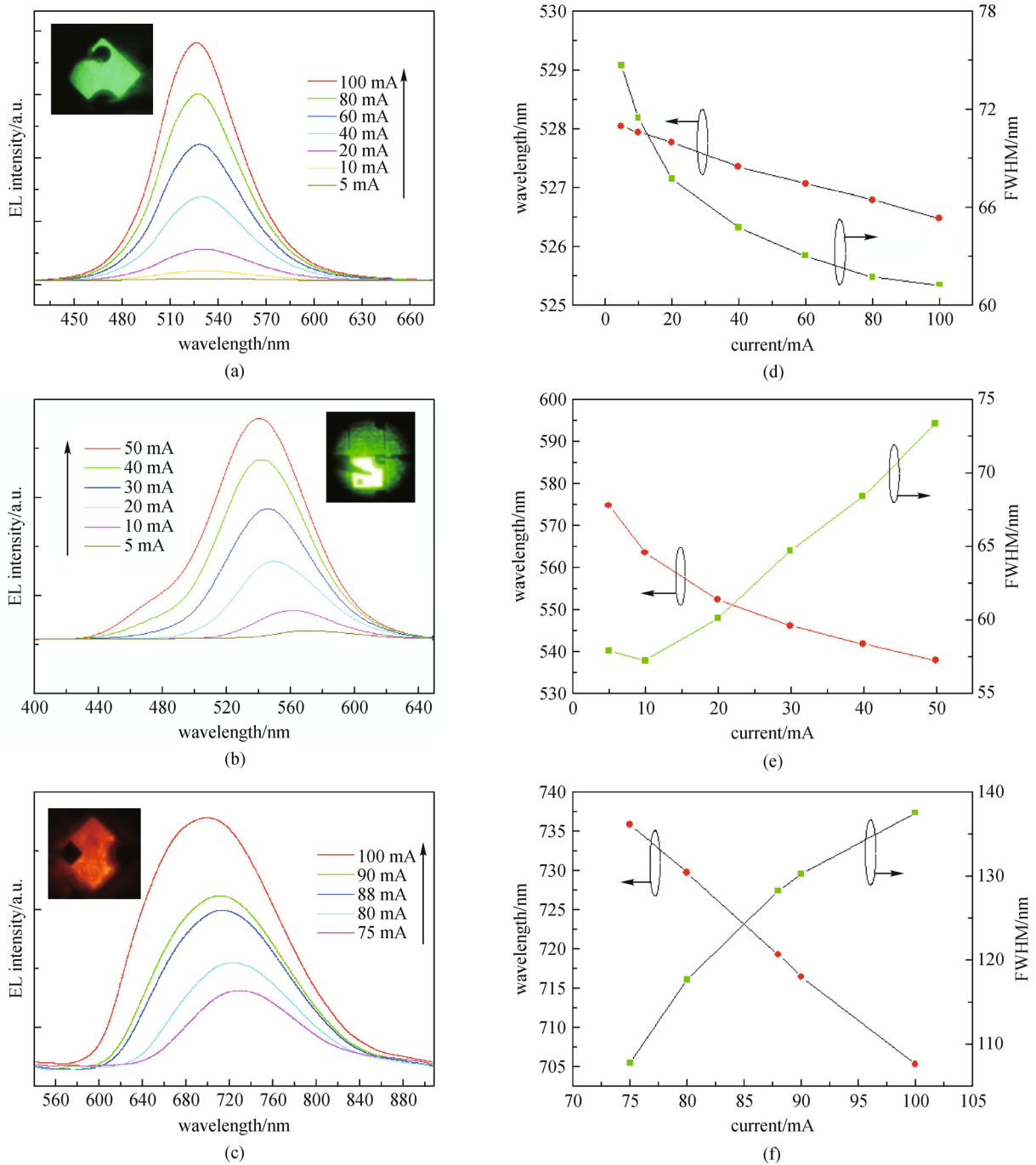


Fig. 6 EL spectra of the green (a) [30], yellow-green (b) [29] and red (c) [26] InGaN QD LEDs under different injection current at room temperature; (d) to (f) show the dependence of peak wavelength and FWHM on injection current density of green, yellow-green and red LEDs, respectively. After W. B. Lv et al. [30]. Reprinted from Applied Physics Express, 7, W.B. Lv, et al., pp. 025203. Copyright (2014), with permission from The Japan Society of Applied Physics. After W. B. Lv et al. [26]. Reprinted from Japanese Journal of Applied

is estimated to be 1 mW at a driving current of 20 mA by on-wafer measurements using a calibrated photodiode. As for the yellow-green InGaN QD LEDs, the light output power is estimated to be 1 mW at a driving current of 50 mA in the same way. In fact, the light output power of deep-red InGaN QD LEDs is even lower than that of green and

yellow-green LEDs. Some measures should be adopted in the epitaxial growth of InGaN QDs, including increasing densities and decreasing aspect ratio. Other measures can be used to improve the performance of InGaN QD LEDs, such as indium tin oxide (ITO) contacts, surface roughening and patterned substrates, etc.

7.3 Phosphor free white LEDs

The color quality of phosphor-free white LEDs is better than blue LEDs plus yellow phosphors. The solution of phosphor-free white LEDs can be realized only by InGaN material system. As for the active region, it may consist of blue InGaN QWs and yellow InGaN QDs. Blue InGaN QWs, GaN barrier layers and yellow InGaN QDs can be grown successively, or blue InGaN QWs plus green and red InGaN QDs, etc. To achieve high color quality and high performance, growth parameters and mechanism should be investigated intensively.

8 Conclusion

Theoretical study reveals that when the indium composition is higher than 27%, dislocations become to generate more easily than QDs. So the growth interruption method is introduced to the growth of high indium composition InGaN QDs. In the growth interruption, the three-dimensional axial strain components are reduced while forming QDs, which can suppress the QCSE to some extent. As for the multilayer QDs, it is important to optimize the growth parameters of GaN barriers. Depending on the growth interruption method and optimized GaN barrier growth situation, green, yellow-green and red LEDs are successfully demonstrated respectively. The developments of long wavelength range QDs and blue QWs LEDs can implement tricolor white LEDs based on only InGaN material system, for the coverage of the entire visible spectrum. More significantly, multilayer InGaN QDs plus QWs served as active region, it paves a potential way to fabricate the phosphor free white LEDs even in a single chip.

Acknowledgements This work was supported by the National Basic Research Program of China (Nos. 2013CB632804, 2011CB301900 and 2012CB3155605), the National Natural Science Foundation of China (Grant Nos. 61176015, 61210014, 51002085, 61321004, 61307024 and 61176059), and the High Technology Research and Development Program of China (Nos. 2011AA03A112, 2011AA03A106, 2011AA03A105 and 2012AA050601).

References

1. Taguchi T. Present status of energy saving technologies and future prospect in white LED lighting. *IEEJ Transactions on Electrical and Electronic Engineering*, 2008, 3(1): 21–26
2. Wang Q, Wang T, Bai J, Cullis A G, Parbrook P J, Ranalli F. Influence of annealing temperature on optical properties of InGaN quantum dot based light emitting diodes. *Applied Physics Letters*, 2008, 93(8): 081915-1–081915-3
3. Jarjour A F, Taylor R A, Oliver R A, Kappers M J, Humphreys C J, Tahraoui A. Cavity-enhanced blue single-photon emission from a single InGaN/GaN quantum dot. *Applied Physics Letters*, 2007, 91(5): 052101-1–052101-3
4. Detchprohm T, Zhu M W, Li Y F, Zhao L, You S, Wetzel C, Preble E A, Paskova T, Hanser D. Wavelength-stable cyan and green light emitting diodes on nonpolar m-plane GaN bulk substrates. *Applied Physics Letters*, 2010, 96(5): 051101-1–051101-3
5. Detchprohm T, Zhu M W, Li Y F, Xia Y, Wetzel C, Preble E A, Liu L H, Paskova T, Hanser D. Green light emitting diodes on a-plane GaN bulk substrates. *Applied Physics Letters*, 2008, 92(24): 241109-1–241109-3
6. Lin Y D, Chakraborty A, Brinkley S, Kuo H C, Melo T, Fujito K, Speck J S, DenBaars S P, Nakamura S. Characterization of blue-green m-plane InGaN light emitting diodes. *Applied Physics Letters*, 2009, 94(26): 261108-1–261108-3
7. Funato M, Ueda M, Kawakami Y, Narukawa Y, Kosugi T, Takahashi M, Mukai T. Blue, green, and amber InGaN/GaN light-emitting diodes on semipolar {11-22} GaN bulk substrates. *Japanese Journal of Applied Physics*, 2006, 45(7L): L659
8. Sato H, Tyagi A, Zhong H, Fellows N, Chung R B, Saito M, Fujito K, Speck J S, DenBaars S P, Nakamura S. High power and high efficiency green light emitting diode on free-standing semipolar (112) bulk GaN substrate. *Physica Status Solidi. Rapid Research Letters*, 2007, 1(4): 162–164
9. Yamamoto S, Zhao Y J, Pan C C, Chung R B, Fujito K, Sonoda J, DenBaars S P, Nakamura S. High-efficiency single-quantum-well green and yellow-green light-emitting diodes on semipolar (20) GaN substrates. *Applied Physics Express*, 2010, 3(12): 122102
10. Feezell D F, Schmidt M C, DenBaars S P, Nakamura S. Development of nonpolar and semipolar InGaN/GaN visible light-emitting diodes. *MRS Bulletin*, 2009, 34(5): 318–323
11. Young E C, Wu F, Romanov A E, Tyagi A, Gallinat C S, DenBaars S P, Nakamura S, Speck J S. Lattice tilt and misfit dislocations in (11) semipolar GaN heteroepitaxy. *Applied Physics Express*, 2010, 3(1): 11004-1–11004-4
12. Craven M D, Lim S H, Wu F, Speck J S, DenBaars S P. Threading dislocation reduction via laterally overgrown nonpolar (1120) a-plane GaN. *Applied Physics Letters*, 2002, 81(7): 1201–1203
13. Masui H, Nakamura S, DenBaars S P, Mishra U K, Ieee T. Nonpolar and semipolar III-nitride light-emitting diodes: achievements and challenges. *IEEE Transactions on Electron Devices*, 2010, 57(1): 88–100
14. Zhao W, Wang L, Wang J X, Hao Z B A, Luo Y. Theoretical study on critical thicknesses of InGaN grown on (0001) GaN. *Journal of Crystal Growth*, 2011, 327(1): 202–204
15. People R, Bean J C. Calculation of critical layer thickness versus lattice mismatch for $\text{Ge}_x\text{Si}_{1-x}/\text{Si}$ strained-layer heterostructures. *Applied Physics Letters*, 1985, 47(3): 322–324
16. Nakajima K. Equilibrium phase diagrams for Stranski-Krastanov structure mode of III–V ternary quantum dots. *Japanese Journal of Applied Physics*, 1999, 38(4R): 1875
17. Zhao W, Wang L, Lv W B, Wang L, Wang J X, Hao Z B, Luo Y. Growth behavior of high-indium-composition InGaN quantum dots using growth interruption method. *Japanese Journal of Applied Physics*, 2011, 50(6R): 065601
18. Yao H H, Lu T C, Huang G S, Chen C Y, Liang W D, Kuo H C, Wang S C. InGaN self-assembled quantum dots grown by metal-organic chemical vapour deposition with growth interruption.

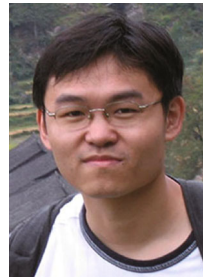
Nanotechnology, 2006, 17(6): 1713–1716

19. Ji L W, Su Y K, Chang S J, Wu L W, Fang T H, Chen J F, Tsai T Y, Xue Q K, Chen S C. Growth of nanoscale InGaN self-assembled quantum dots. *Journal of Crystal Growth*, 2003, 249(1–2): 144–148
20. Ee Y K, Zhao H P, Arif R A, Jamil M, Tansu N. Self-assembled InGaN quantum dots on GaN emitting at 520 nm grown by metalorganic vapor-phase epitaxy. *Journal of Crystal Growth*, 2008, 310(7–9): 2320–2325
21. Bayram C, Razeghi M. Density-controlled growth and field emission property of aligned ZnO nanorod arrays. *Applied Physics A*, 2009, 97(2): 403–408
22. Ji L W, Su Y K, Chang S J, Wu L W, Fang T H, Xue Q K, Lai W C, Chiou Y Z. A novel method to realize InGaN self-assembled quantum dots by metalorganic chemical vapor deposition. *Materials Letters*, 2003, 57(26–27): 4218–4221
23. Wang Q, Wang T, Parbrook P J, Bai J, Cullis A G. The influence of a capping layer on optical properties of self-assembled InGaN quantum dots. *Journal of Applied Physics*, 2007, 101(11): 113520-1–113520-7
24. Wang Q, Wang T, Bai J, Cullis A G, Parbrook P J, Ranalli F. Growth and optical investigation of self-assembled InGaN quantum dots on a GaN surface using a high temperature AlN buffer. *Journal of Applied Physics*, 2008, 103(12): 123522-1–123522-4
25. Bai J, Wang Q, Wang T, Cullis A G, Parbrook P J. Optical and microstructural study of a single layer of InGaN quantum dots. *Journal of Applied Physics*, 2009, 105(5): 53505-1–53505-5
26. Lv W B, Wang L, Wang J X, Xing Y C, Zheng J Y, Yang D, Hao Z B, Luo Y. Green and red light-emitting diodes based on multilayer InGaN/GaN dots grown by growth interruption method. *Japanese Journal of Applied Physics*, 2013, 52(8S): 08JG13-1–08JG13-2
27. Wang L, Zhao W, Lv W B, Wang L, Hao Z B, Luo Y. The influence of underlying layer on morphology of InGaN quantum dots self-assembled by metal organic vapor phase epitaxy. *Physica Status Solidi C*, 2012, 9(3–4): 782–785
28. Lv W B, Wang L, Wang J, Hao Z, Luo Y. Density increase of upper quantum dots in dual InGaN quantum-dot layers. *Chinese Physics Letters*, 2011, 28(12): 128101-1–128101-3
29. Lv W B, Wang L, Wang J, Hao Z, Luo Y. InGaN/GaN multilayer quantum dots yellow-green light-emitting diode with optimized GaN barriers. *Nanoscale Research Letters*, 2012, 7(1): 617–624
30. Lv W B, Wang L, Wang L, Xing Y C, Yang D, Hao Z B, Luo Y. InGaN quantum dot green light-emitting diodes with negligible blue shift of electroluminescence peak wavelength. *Applied Physics Express*, 2014, 7(2): 025203



and sensors.

Lai Wang received his BS and PhD degrees from Tsinghua University in 2003 and 2008, respectively, all in Electronic Engineering. Associate Prof. Lai Wang's current research interests include: MOCVD growth and characterization of III-nitride materials and their applications; carrier dynamics and efficiency droop in InGaN MQWs; InGaN QDs and light-emitting devices; detectors



Wenbin Lv received his BS and MS degrees from Dalian Maritime University in 2002 and 2005, respectively, all in Communication Engineering. Now he is a PhD candidate in Department of Electronic Engineering, Tsinghua University. His current research interests include InGaN QDs and its light-emitting devices.



Zhibiao Hao received his BS and PhD degrees from Tsinghua University in 1996 and 2001, respectively, all in Electronic Engineering. Prof. Zhibiao Hao's current research interests include: epitaxial growth of III-V compound semiconductors; nanostructure optoelectronic devices; wide band-gap electronic devices; fundamentals of microwave and optical wave.



Yi Luo received his BS from Tsinghua University and his MS and PhD degrees from University of Tokyo in 1983, 1987 and 1990, respectively, all in Electronic Engineering. Prof. Yi Luo's current research interests include: fabrication and package technologies of photonic integrated devices for fiber telecommunication and radio over fiber system; materials growth, device fabrication and optical system design for LED-based solid state lighting; novel photonic devices based on nanostructure; solar cell and solar thermal application technology.