High-color-rendering-index phosphor-free InGaN-based white light-emitting diodes by carrier injection enhancement via V-pits

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ABSTRACT
We herein report the growth of phosphor-free InGaN-based white light-emitting diodes (LEDs) by metalorganic vapor-phase epitaxy. The active region consists of blue and red InGaN quantum wells (QWs). To improve the current injection and generate broadband emission, the V-pit structures in the LEDs were fabricated intentionally before growing the QWs. The monolithic white LEDs emit in the range of 410–770 nm and, by tuning the injection current, can cover correlated color temperature (CCT) values corresponding to warm white, natural white, and cool white. The color-rendering index (CRI) of the white LEDs reaches 88 at an injection current of 10 mA. At an injection current of 30 mA, the white LEDs exhibit the chromaticity coordinates of (0.320 and 0.334) in the Commission Internationale de l’Eclairage 1931 chromaticity diagram, a CRI of 78, and a CCT of 6110 K.

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Commercial white light-emitting diodes (LEDs) couple InGaN-based blue or violet LEDs with wavelength-converting phosphors, which is the conventional method for achieving white emission." The blue or violet LEDs serve to optically pump the phosphors, converting part of the LED emission to longer wavelengths via phosphor re-emission. However, this method has several disadvantages, such as Stokes-shift energy loss and phosphor degradation. Therefore, the fabrication of phosphor-free white LEDs is highly desirable.

By tuning the alloy composition of III-nitride materials, they can be made to emit over the full visible spectrum, which makes them excellent candidates for monolithically integrated white LEDs with polychromatic emission. Researchers have proposed several approaches to produce monolithic, phosphor-free InGaN-based white LEDs, such as multiple quantum wells (MQWs) with different emission wavelengths, nanostructured array structures, quantum dot structures, patterned microstructures, and optically pumped QWs. In particular, LED fabrication requires a simplified process that does not involve phosphors or patterned nano or microstructures. However, LEDs with monolithically embedded multicolor planar QWs require high current injection (>200 mA) to obtain white emission. InGaN MQWs with various In contents can emit polychromatic light, which, under certain conditions, can result in white emission. However, a disadvantage of these white LEDs remains the difficult hole injection in the planar MQWs across the growth direction. A nonuniform carrier distribution across InGaN QWs is caused by the low hole mobility and a piezoelectric field. Therefore, LEDs should be designed to enhance the hole injection by, such as, QW stacking order, V-pits, and polarization.

In general, typical monolithic InGaN-based white LEDs lack the red emission component, which renders the color rendering index (CRI) insufficient (e.g., <70) for white emission. To emit at longer wavelengths, InGaN requires a high-In-content, but obtaining the requisite In content in a high-quality crystal represents a significant challenge. Growing high-In-content InGaN presents several problems, including low-temperature growth, a large lattice mismatch with GaN, and the quantum-confined Stark effect (QCSE). Thus, the QWs of red InGaN LEDs require further improvement before they can be used in highly efficient devices.

In this study, we demonstrate phosphor-free white LEDs based on integrated InGaN QWs with blue and red emissions. We obtained red InGaN QWs by using our recently developed InGaN-based red-LED technologies such as high-temperature growth, strain
engineering, and hybrid MQW structures. In addition, the V-pit structure is vital for injecting holes into the QWs. V-pits are introduced into the LED structures to improve hole injection into the blue and red QWs. The broad spectral emission translates into a high CRI value, which, by varying the injection current, can be tuned to the correlated color temperature (CCT) of different shades of white, such as cool white or warm white.

The LED structures were grown by metalorganic vapor-phase epitaxy in a single-wafer horizontal reactor. To improve the light-extraction efficiency, we used c-plane sapphire substrates with a cone-shaped patterned structure. The red-emitting QW structure consisted of AlN/AlGaN barrier layers to compensate for strain and, thereby, improve the crystal quality. Figure 1 shows a schematic cross-sectional view of the white LED structure. The LEDs consist of an unintentionally doped (uid) GaN layer (2 μm) with a low-temperature GaN buffer layer, an n-GaN:Si layer (8 μm, n = 2 × 10^{18} \text{ cm}^{-3}), an n-Al_{0.13}Ga_{0.87}N (18 nm)/GaN (3 nm) barrier layers, 15 periods of uid-GaN (6 nm)/uid-In_{0.08}Ga_{0.92}N (2 nm) superlattices (SLs), an n-GaN:Si layer (15 nm), a blue single-quantum well (SQW), red double-quantum wells (DQWs), an uid-GaN layer (15 nm), a p-GaN:Mg layer (100 nm), and a p^+ -GaN:Mg contact layer (10 nm). The QW structures consist of an In_{0.2}Ga_{0.8}N blue SQW (2 nm) with GaN (2 nm)/Al_{0.13}Ga_{0.87}N (18 nm)/GaN (3 nm) barrier layers, and In_{0.13}Ga_{0.87}N red DQWs (2.5 nm) with AlN (12.5 nm)/GaN (2 nm)/ Al_{0.13}Ga_{0.87}N (18 nm)/GaN (3 nm) barrier layers. GaN was used to replace the Al_{0.13}Ga_{0.87}N part in the upper barrier of the second red QW. During the n-AlGaN growth, V-pits form in the LED structures, and the n-AlGaN layer has a low resistivity due to its high concentration of Si dopants. The 250 × 650 μm² LED chips were fabricated by using the standard face-up mesa method. A 120-nm-thick indium tin oxide layer was deposited on the LED surface by sputtering, following which mesa-etching was carried out by inductively coupled plasma etching to expose the n-AlGaN layer. Cr (50 nm)/Ni (20 nm)/Au (200 nm) were deposited as n- and p-pad electrodes by e-beam deposition.

The LED structure was imaged by scanning electron microscopy (SEM) and scanning transmission electron microscopy (STEM) before fabricating the LED devices. The electroluminescence (EL) was measured directly on-wafer, under direct current (DC) operation at room temperature (RT).

Figure 2(a) shows SEM images of the surface morphology of the LEDs and reveals several V-pits of two different sizes on the LED surface: large V-pits approximately 500 nm in diameter and small V-pits approximately 100 nm in diameter. The large-V-pit density is 1.5 × 10^8 \text{ cm}^{-2}, which corresponds to the dislocation density of the underlying GaN layer. Cross-sectional STEM observation indicates that these large V-pits are generated from the n-AlGaN layers via threading dislocations, as shown in Fig. 2(b). The V-pits form on the {1011} facets, which is a typical plane of Ga-polar growth and has the potential of improving hole injection into the QWs. Also, the semi-polar sidewall QWs should have a reduced In content and QCSE, which should enhance the emission at wavelengths below those emitted by the regular sections of QWs. The improved current injection and emission from the semi-polar sidewall QWs are significant for obtaining monolithic white LEDs. The small V-pits are introduced by the degradation of the red DQWs, which is attributed to the fact that defects in high-In-content InGaN QWs are easily introduced at low growth temperature and lattice mismatch.

Figure 3(a) shows EL spectra for current injection ranging from 5 to 100 mA. The LEDs emit over a wide range for all current levels measured. The emission is dominated by two components, blue and red, which is due to the dichromatic QWs. Both emissions blueshift as the current injection increases. The blue and red peaks blueshift by 16 and 22 nm, respectively, as the current injection increases from 5 to 100 mA. These large blueshifts are typical and are attributed to the screening by the QCSE. The localized emissions are distributed in the LED chips, which contributes to broadening the EL emission, as shown in Fig. 3(b). The high-In-content InGaN LEDs have localized states due to phase separation. Also, the localized blue emission comes from the QWs in the semi-polar sidewalls of the V-pits, and the mixing of these localized emissions improves the CRI value for a natural white emission, as shown in Fig. 3(c). In contrast, LEDs without the large V-pits only produce red emission, as discussed in our

**FIG. 1.** Schematic of the cross-sectional view of the epi-structure of monolithic InGaN-based white LEDs.

**FIG. 2.** (a) Plan-view SEM and (b) cross-sectional STEM images of white LEDs.
Therefore, V-pits contribute significantly to the injection current into the blue SQW.

Figure 4 shows the CRI value and the blue/red peak ratio as a function of injection current. The CRI value of the monolithic white LED reaches as high as 88 at an injection current of 10 mA. The CRI value is sensitive to the blue/red peak ratio, and the results show that the CRI decreases as the blue/red peak ratio increases. Nevertheless, the CRI value remains as high as 73 at high current injection, which is attributed to the red emission component. Upon exceeding a current injection of 20 mA, the main emission peak switches from red to blue, which can contribute to the color tunability of the white emission.

Figure 5 shows Commission Internationale de l’Eclairage (CIE) 1931 chromaticity coordinates corresponding to the EL spectra of the white LEDs at various current-injection levels. The LEDs can be tuned from warm white to cool white emission. At a low injection current of 5 mA, the EL emission is in the warm white range; as the current increases to 100 mA, the blue emission component is significantly enhanced, and the EL emission shifts to the cool-white range. Notably, the EL emission at 30 mA obtained the CIE coordinates \((x, y) = (0.320, 0.334)\), which is close to the white point of \((0.3127, 0.3290)\) in Rec. 2020. This white emission corresponds to a CRI of 78 and follows closely to the Planckian locus. The CCT also shifts gradually and attains 3150, 3720, 6110, and 12 600 K at 5, 10, 30, and 100 mA, respectively.

Figure 6 shows the forward voltage and luminous flux characteristics of the white LEDs at various DC injection levels and at RT. The forward voltage for the white LEDs is 3.0 V at 20 mA, which is less than the value obtained from our previous red LEDs (3.4 V at 20 mA). The lower forward voltage in the white LEDs is attributed to enhanced carrier injection via V-pits. The luminous flux increases linearly with current injection. Finally, the electrical characteristics of the device indicate a good p–n junction for monolithic white LEDs.

In summary, we demonstrate in this work white LEDs with monolithically integrated QWs. The white LED structure was fabricated via a simplified phosphor-free process involving no patterned nano or microstructures. The formation of large V-pits in the n-AlGaN layers contributes to carrier transport into the blue and red QWs. The monolithic white LEDs produce a CRI as high as 88 at an injection current of 10 mA (2.8 V). They also emit high-quality white light with CIE \((x, y)\) chromaticity coordinates of \((0.320, 0.334)\), a CRI of 78, and a CCT of 6110 K at 30 mA (3.2 V). By varying the current injection, the LED emission color is tunable from warm white to cool...
white. Thus, these monolithic white LEDs are promising devices for the development of phosphor-free white-light sources.

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DATA AVAILABILITY

The data that support the findings of this study are available within this article.

REFERENCES

1. E. F. Schubert and J. K. Kim, Science 308, 1274 (2005).
2. Y. Narukawa, M. Ichikawa, D. Sanga, M. Sano, and T. Mukai, J. Phys. D 43, 354002 (2010).
3. K. W. Houser, M. Wei, A. David, and M. R. Krames, Leukos 10, 165 (2014).
4. M. Meneghini, L.-R. Trevisanello, F. de Zuanii, N. Trivellini, G. Meneghesso, and E. Zanoni, Proc. SPIE 7422, 74220H (2009).
5. M. Yamada, Y. Narukawa, and T. Mukai, Jpn. J. Appl. Phys., Part 2 41, L246 (2002).
6. K. Lee, I. Choi, C. R. Lee, T. H. Chung, Y.-S. Kim, K.-U. Jeong, D.-C. Chung, and J. S. Kim, Sci. Rep. 7, 7164 (2017).
7. A. Yadav, I. E. Titkov, A. V. Sarkharov, W. V. Lundin, A. E. Nikolaev, G. S. Sokolovskii, A. F. Tsatsulnikov, and E. U. Rafailov, Appl. Phys. Lett. 118 (2018).
8. N. Poyiatzis, M. Athanasiou, J. Bai, Y. Gong, and T. Wang, Sci. Rep. 9, 1383 (2019).
9. W. Lin, Y. J. Lu, H. Y. Chen, H. M. Lee, and S. Gwo, Appl. Phys. Lett. 97, 033101 (2010).
10. M. R. Philip, D. D. Choudhary, M. Djavid, M. N. Bhuyian, J. Piao, T. T. Pham, D. Misra, and H. P. T. Nguyen, J. Vac. Sci. Technol., B 35, 02B108 (2017).
11. K. Kishino, N. Sakakibara, K. Narita, and T. Oto, Appl. Phys. Express 13, 014003 (2020).
12. H. J. Li, P. P. Li, J. J. Kang, Z. Li, L. Z. Li, L. J. Li, X. Y. Yi, and G. H. Wang, Appl. Phys. Express 6, 102103 (2013).
13. H. Li, P. Li, J. Kang, J. Ding, J. Ma, Y. Zhang, X. Yi, and G. Wang, Sci. Rep. 6, 35217 (2016).
14. C. P. Cho, I. K. Park, M. K. Kwon, J. Y. Kim, S. J. Park, D. R. Jung, and K. W. Kwon, Appl. Phys. Lett. 93, 241109 (2008).
15. M. Funato, T. Kondou, K. Hayashi, S. Nishiura, M. Ueda, Y. Kawakami, Y. Narukawa, and T. Mukai, Appl. Phys. Express 1, 011006 (2008).
16. M. L. Lee, Y. H. Yeh, S. J. Tu, P. C. Chen, W. C. Lai, and J. K. Sheu, Opt. Express 23, A401 (2015).
17. G. F. Yang, P. Chen, S. M. Gao, G. Q. Chen, R. Zhang, and Y. D. Zheng, Photonics Res. 4, 17 (2016).
18. B. Damilano, P. Demolom, J. Brault, T. Huaulf, F. Natali, and J. Massies, J. Appl. Phys. 108, 073115 (2010).
19. S. J. Kowcz, C. D. Pynn, S. H. Oh, R. M. Farrell, J. S. Speck, S. P. DenBaars, and S. Nakamura, Appl. Phys. Lett. 107, 101104 (2015).
20. D. Kang, J.-T. Oh, J.-O. Song, T.-Y. Seong, M. Keinsell, and H. Amano, Appl. Phys. Express 12, 102016 (2019).
21. A. Kosikin, N. Takahashi, T. Taki, and H. Seki, J. Cryst. Growth 170, 306 (1997).
22. Y. Yamashita, H. Tamura, N. Horio, H. Sato, K. Taniguchi, T. Chinone, S. Omori, and C. Funakita, Jpn. J. Appl. Phys., Part 1 42, 4197 (2003).
23. M. Shimizu, Y. Kawaguchi, K. Hiramatsu, and N. Sawaki, Jpn. J. Appl. Phys., Part 1 36, 3381 (1997).
24. D. Holec, P. M. F. J. Costa, M. J. Kappers, and C. J. Humphreys, J. Cryst. Growth 303, 314 (2007).
25. T. Takeuchi, S. Sota, M. Katsuragawa, M. Komori, H. Takeuchi, H. Amano, and I. Akasaki, Jpn. J. Appl. Phys., Part 2 36, L382 (1997).
26. T. Takeuchi, C. Wetzel, S. Yamaguchi, H. Sakai, H. Amano, I. Akasaki, Y. Kaneko, S. Nakagawa, Y. Yamaoka, and N. Yamada, Appl. Phys. Lett. 73, 1691 (1998).
27. K. Ohkawa, T. Watanabe, M. Sakamoto, A. Hirako, and M. Deura, J. Cryst. Growth 343, 13 (2012).
28. K. Ohkawa, F. Ichinohe, T. Watanabe, K. Nakamura, and D. Iida, J. Cryst. Growth 512, 69 (2019).
29. D. Iida, S. Lu, S. Hirahara, K. Niwa, S. Kamiyama, and K. Ohkawa, J. Cryst. Growth 448, 105 (2016).
30. D. Iida, Z. Zhuang, P. Kirilenko, M. Velazquez-Rizo, M. A. Najmi, and K. Ohkawa, Appl. Phys. Lett. 116, 162101 (2020).
31. D. Iida, K. Niwa, S. Kamiyama, and K. Ohkawa, Appl. Phys. Express 9, 111003 (2016).
32. Q. Lv, J. Liu, C. Mo, J. Zhang, X. Wu, Q. Wu, and F. Jiang, ACS Photonics 6, 130 (2019).
33. F. Jiang, I. Zhang, L. Xu, J. Ding, G. Wang, X. Wu, X. Wang, C. Mo, Z. Quan, X. Guo, C. Zheng, S. Pan, and J. Liu, Photonics Res. 7, 144 (2019).
34. D. Iida, Z. Zhuang, P. Kirilenko, M. Velazquez-Rizo, and K. Ohkawa, Appl. Phys. Express 13, 031001 (2020).
35. T. Sugiyama, D. Iida, T. Yasuda, M. Iwaya, T. Takeuchi, S. Kamiyama, and I. Akasaki, Appl. Phys. Express 6, 121002 (2013).
36. Z. Zhuang, D. Iida, P. Kirilenko, M. Velazquez-Rizo, and K. Ohkawa, Opt. Express 28, 12311 (2020).
37. Y. Wang, R. Shimma, T. Yamamoto, H. Hayashi, K.-I. Shiohama, K. Kurihara, R. Hasegawa, and K. Ohkawa, J. Cryst. Growth 416, 164 (2015).
38. W. R. Zhao, G. E. Weng, J. Y. Wang, J. Y. Zhang, H. W. Liang, T. Sekiguchi, and B. P. Zhang, Nanoscale Res. Lett. 10, 459 (2015).
39. J.-I. Hwang, R. Hashimoto, S. Saito, and S. Nunoue, Appl. Phys. Express 7, 071003 (2014).