Control performance of a single-chip white light emitting diode by adjusting strain in InGaN underlying layer

X. H. Wang, L. W. Guo,^{a)} H. Q. Jia, Z. G. Xing, Y. Wang, X. J. Pei, J. M. Zhou, and H. Chen

Beijing National Laboratory of Condensed Matter, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, People's Republic of China

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Light emission from green to white in a single-chip light emitting diode is modulated by adjusting the strain in InGaN underlying layer (UL) embedded below an active layer of InGaN/GaN multiple quantum wells. Transmission electron microscopy combined with x-ray reciprocal space mapping reveals that indium phase separation in InGaN quantum well active layer is enhanced by using a partly relaxed InGaN UL and In-rich quantum dots with different size and indium composition are formed. They emit multicolor lights whose mixing produces white light. Quality of the white light could be controlled by modulation on relaxation degree of the InGaN UL. © 2009 American Institute of Physics. [DOI: 10.1063/1.3103559]

Recent progress on light emitting diodes (LEDs) pushes LED as a strong competitive candidate for lighting applications,¹⁻⁴ owing to its long lifetime, small size, and low energy consumption. At present, the most commonly used method to achieve a white LED is to combine a blue GaN based LED chip with yttrium aluminum garnet phosphor that emits yellow light.⁵ However, there are problems with this type of white LED, such as high color temperature and low color rendering index (CRI).⁶ Moreover, package process of this type of white LED is complicated. Meanwhile, there are also limitations in making white LED without phosphor converter, including photon recycled white LED,⁷ white LED with codoped active well layer,⁸ and a combination of two InGaN/GaN LEDs emitted different colors.⁹ The limitations are low light emission efficiency in photo recycled LED, saturated light emission and poor white light quality in the codoped method, and complicated device processes and circuit control in the double colors LEDs combination. Therefore, fabricating phosphor-free white LED with simple processes has been an objective of researchers engaged in the field. In 2007, single-chip white LEDs were fabricated successfully in our laboratory with white light emitting from InGaN/GaN multiple quantum wells (MQWs) active layer grown on a thick InGaN underlying layer (UL). Their emission intensity did not show saturation with increasing injected current.¹⁰ To reveal correlation of the white LED performance with the InGaN UL, a dependence of white light quality on the strain of the InGaN UL is studied in this work. Three InGaN ULs with different thicknesses are designed, on which 4 periods InGaN MQWs were grown with same growth parameters and conditions. It is found that there exist multi-peak emissions from the LEDs, whose mixing gives white light in sight view. Meanwhile, a possible light emission mechanism of the single-chip white LED is revealed.

The samples were grown on (0001)-oriented sapphire substrates by using low-pressure metalorganic vapor phase epitaxy. The strain of InGaN UL was adjusted by changing the thickness of the InGaN UL while keeping indium composition, on which same InGaN/GaN QWs were grown. The structure of our LED is composed of a 30-nm-thick GaN nucleation layer, a 3- μ m-thick Si-doped GaN layer, followed by an InGaN UL. Then active layer of four periods InGaN (3 nm)/GaN (14 nm) MQWs was deposited. At last, a 0.2- μ m-thick Mg-doped *p*-GaN was grown. Thicknesses of InGaN ULs in samples A, B, and C are 160, 190, and 220 nm, respectively. LED chips with the size of 0.3 ×0.3 mm² were processed by photolithography and etching. TiAl and NiAu alloys were used for *n*- and *p*-type Ohmic contacts, respectively.

Relaxation degree of InGaN ULs and their indium compositions were studied by asymmetrical (105) x-ray reciprocal space mapping (RSM). Electroluminescence (EL) measurements were performed in analyzing quality of the white light emission. Microstructures of the InGaN MQWs were studied by transmission electron microscopy (TEM).

Figure 1 shows the EL spectra of the samples with 7 mA injection current. At this injection current, at least two clearly separated emission peaks from InGaN MQWs of the samples are observed. By fitting experimental curves with Gaussian functions, possible emission peaks on the samples are shown. In sample A, there are two clearly separated peaks. One is located at 436 nm that is weak and the other is at 545 nm that is strong. However, three clear peaks are shown in samples B and C. They are located at 441, 494, and 563 nm in sample B and 443, 496, and 576 nm in sample C. It should be emphasized that in order to reveal more emission peaks a 7 mA current was injected to the LEDs instead of commonly used 20 mA. As a higher injection current is used, the relative weak fitting peak at middle wavelength in samples B and C is emerged into the strong background of the short and the long wavelength peaks. Intensity ratios of the short wavelength bluelike emission peak to the long wavelength emission peak from the samples A, B, and C are 0.03, 0.17, and 0.52, respectively. So, sight view of sample A is green, sample B is yellow-green, and sample C is white. Clearly, completely different EL view colors are observed from the samples with different thicknesses of InGaN UL. It

^{a)}Author to whom correspondence should be addressed. Electronic mail: lwguo@aphy.iphy.ac.cn.

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FIG. 1. (Color online) EL spectra of (a) sample A, peaked at 436 and 545 nm, (b) sample B, peaked at 441, 494, and 563 nm, and (c) sample C, peaked at 443, 496, and 576 nm.

reveals a close relation between light emissions and InGaN ULs.

Therefore, origins of multipeak emissions from InGaN MQW active layer and peak intensity ratios were studied. Asymmetrical x-ray RSM of the (105) reflection was measured, as shown in Fig. 2. It is observed that relative posi-



FIG. 2. X-ray RSM of the (105) reflection for (a) sample A, (b) sample B, and (c) sample C. Two diffraction peaks correspond to GaN and InGaN UL, respectively. Dashed lines guide diffraction peak positions of the completely relaxed and strained InGaN alloys. (c) is cited from Ref. 10.

tions between InGaN UL diffraction peaks to GaN reference peak are different in three samples. Conditions and parameters used in growing InGaN UL are the same except InGaN UL thicknesses, which induces different strain status in In-GaN UL. Indium composition and strain (or relaxation degree) of the InGaN ULs in three samples were deduced in Fig. 2 combined with Poisson ratio and relaxation degree definition of a layer with given composition.^{11–13} The deduced indium composition is $4.4\% \pm 0.4\%$ in all samples and relaxation degrees are about 9.6%, 31.8% and 64.4% for samples A, B, and C, respectively.^{12,13}

In general, phase separation of indium in InGaN alloy is suppressed by biaxial strain.^{13,14} In our experiments, InGaN MQWs were grown on a partly relaxed InGaN UL with low indium composition. Taking the partly relaxed InGaN UL as a strain-adjusting template, InGaN MQWs grown on it suffer from a relative weak strain compared with that grown on GaN directly. In this case, enhanced phase separation of indium or accumulation of indium in InGaN MQWs takes place especially in samples B and C.

Luminescence from InGaN/GaN MQWs LED is considered relating to In-rich quantum dots (QDs) or potential minima existed in InGaN QW layers.^{15,16} With correlation between strain of the InGaN UL and intensity of EL as well as sight view colors of EL emissions from the samples, it could be inferred that the In-rich QDs with different lateral size and indium composition are distributed in the same In-GaN active layer. Their sizes, densities, and indium compositions in QDs have close relations with strain or strain fluctuation on the surface of the InGaN UL. The larger the relaxation degree is the easier the forming of In-rich QDs, which results in further indium accumulation and forming larger size In-rich QDs.^{12,13} In that case, QD density with long wavelength emission is reduced and more injected carriers fill in localized states relating to short wavelength emission. Thereby, multipeak emissions are observed especially in samples B and C, where two long wavelength emissions are supposed from In-rich QDs with different lateral sizes or indium composition, and the short wavelength bluelike emission is from the InGaN QW matrix.^{17,18} In addition, it is noted that EL intensity of samples decreases with increasing InGaN UL thickness, as shown in Fig. 1. It is supposed that dislocations introduced with InGaN UL relaxation in thicker InGaN ULs act as nonradiation recombination centers, trapping injected carriers and lead EL intensity decreased.

To confirm our inference about In-rich QDs in MQWs, cross section TEM images were taken to investigate detail structure of the InGaN/GaN MQWs. The GaN barriers and InGaN wells of the MQWs are shown clearly in Fig. 3. Furthermore, distinguished details in MQWs were observed clearly in TEM images. Dark dots are indistinct in InGaN MQWs of sample A, while numerous dark dots are observed in MQWs of samples B and C, especially in sample C. The TEM images were taken in an enough short time to avoid electron beam induced damage on samples.¹⁷ These dark dots are identified as In-rich QDs. Lateral sizes of these dots are about 1 and 2 nm in sample B and 3 and 4 nm in sample C. QD densities are estimated about 4×10^{12} cm⁻² in sample B and 1×10^{12} cm⁻² in sample C. In Fig. 3(a), a contrast fluctuation in MQWs is faintly discernible, which is considered related to fluctuation of indium composition. Fluctuation of indium composition produces localized potential minima relative to its periphery, which leads to two emission

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FIG. 3. Cross-sectional TEM images of InGaN MQWs active layer grown on different InGaN ULs. They are (a) sample A, (b) sample B, and (c) sample C. In (b) and (c) In-rich QDs are seen clearly.

peaks, as shown in Fig. 1(a), where the 545 nm emission is supposed from local In-rich fluctuations and the 436 nm emission is from the QW matrix. Local fluctuation of indium composition in sample A is in such a small scale that the QD density was hard to evaluate. Therefore, strong long wavelength emission accompanied by weak short wavelength emission was observed as a 7 mA current was injected to the active layer, in which injected carriers are not enough to saturate energy states of the long wavelength. While in samples B and C, part of the carriers could be captured by QW matrix, which produces a short wavelength emission around 441 nm (or 443 nm) as observed in sample B (or C). With increasing injection current, the short wavelength emission intensity increases rapidly, while the long wavelength is nearly unchanged, as discussed in our previous work.¹⁰ The CRI of the white light from sample C reaches a maximum of 53.4 at a current 14 mA and decreased gradually to 36.7 as increasing current to 60 mA. This is mainly attributed to the lack of long wavelength emission with large injecting current.

Based on the experimental observation and discussions above, we suggest that mechanism of multipeak emissions in the LEDs is ascribed to In-rich QDs with different sizes or different indium compositions situated in InGaN MQWs grown on partly relaxed InGaN UL. Degree of indium phase separation or accumulation in InGaN MQWs could be modulated by strain or relaxation degree of the InGaN UL. A better balance on different wavelength emissions is observed in sample C, which is ascribed to large relaxation degree of the InGaN UL in the sample C. At present, we have not accumulated enough evidence to give a clear description on how In-rich QDs with different indium compositions are distributed in InGaN MQWs, which need to be studied further.

In summary, multipeak emissions from same InGaN QW layers are observed in InGaN/GaN MQWs grown on a partly relaxed InGaN strain template in realizing single-chip white LED. Quality of the white light can be modulated by adjusting strain of embedded InGaN UL template. It is found that the higher the relaxation degree of the InGaN UL is the easier the phase separation of indium in InGaN QW layer. And larger In-rich QDs are formed accompanied with a reduced QD density, which leads to a better balance of multicolor emissions. In a well-controlled case, perfect white light emitted from same MQW layer will be realized.

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- ¹S. Nakamura, M. Senoh, S. Nagahama, N. Iwasa, T. Yamada, T. Mukai, Y. Sugimoto, and H. Kiyoku, Appl. Phys. Lett. **69**, 4056 (1996).
- ²S. Nakamura and G. Fasol, *The Blue Laser Diode* (Springer, New York, 1997).
- ³S. Nakamura, IEEE J. Sel. Top. Quantum Electron. **3**, 435 (1997).
- ⁴E. Woelk, G. Strauch, D. Schmitz, M. Deschler, and H. Jurgensen, Mater. Sci. Eng., B **44**, 419 (1997).
- ⁵B. Damilano, N. Grandjean, C. Pernot, and J. Massies, Jpn. J. Appl. Phys., Part 2 **40**, L918 (2001).
- ⁶H.-Y. Chou, T.-H. Hsu, and T.-H. Yang, Proc. SPIE **5739**, 33 (2005).
- ⁷X. Guo, J. W. Graff, and E. F. Schubert, Proc. SPIE **3938**, 60 (2000).
- ⁸J. K. Sheu, C. J. Pan, G. C. Chi, C. H. Kuo, L. W. Wu, C. H. Chen, S. J.
- Chang, and Y. K. Su, IEEE Photonics Technol. Lett. 14, 450 (2002).
- ⁹I. Ozden, E. Makarona, A. V. Nurmikko, T. Takeuchi, and M. Krames, Appl. Phys. Lett. **79**, 2532 (2001).
- ¹⁰X. H. Wang, H. Q. Jia, L. W. Guo, Z. G. Xing, Y. Wang, X. J. Pei, J. M. Zhou, and H. Chen, Appl. Phys. Lett. **91**, 161912 (2007).
- ¹¹A. Tabata, L. K. Teles, L. M. R. Scolfaro, L. R. Leite, A. Kharchenko, T. Frey, D. J. As, D. Schikora, K. Lischika, J. Furthmuller, and F. Bechstedt, Appl. Phys. Lett. **80**, 769 (2002).
- ¹²S. Pereira, M. R. Correia, E. Pereira, K. P. O'Donnell, E. Alves, A. D. Sequeira, and N. Franco, Appl. Phys. Lett. **79**, 1432 (2001).
- ¹³S. Pereira, M. R. Correia, E. Pereira, K. P. O'Donnell, E. Alves, A. D. Sequeira, N. Franco, I. M. Watson, and C. J. Deatcher, Appl. Phys. Lett. **80**, 3913 (2002).
- ¹⁴R. People and J. C. Bean, Appl. Phys. Lett. **47**, 322 (1985).
- ¹⁵S. Chichibu, T. Azuhata, T. Sota, and S. Nakamura, Appl. Phys. Lett. **70**, 21 (1997).
- ¹⁶Y.-L. Lai, C.-P. Liu, Y.-H. Lin, R.-M. Lin, D.-Y. Lyu, Z.-X. Peng, and T.-Y. Lin, Appl. Phys. Lett. **89**, 151906 (2006).
- ¹⁷Y.-L. Lai, C.-P. Liu, and Z.-Q. Chen, Appl. Phys. Lett. 86, 121915 (2005).
- ¹⁸I.-K. Park, M.-K. Kwon, S.-H. Baek, Y.-W. Ok, Y.-S. Kim, Y.-T. Moon, and D.-J. Kim, Appl. Phys. Lett. 87, 061906 (2005).