Effects of multiple interruptions with trimethylindium-treatment in the InGaN/GaN quantum well on green light emitting diodes*

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(Received 21 April 2016; revised manuscript received 25 June 2016; published online 25 August 2016)

In this study, the influence of multiple interruptions with trimethylindium (TMIn)-treatment in InGaN/GaN multiple quantum wells (MQWs) on green light-emitting diode (LED) is investigated. A comparison of conventional LEDs with the one fabricated with our method shows that the latter has better optical properties. Photoluminescence (PL) full-width at half maximum (FWHM) is reduced, light output power is much higher and the blue shift of electroluminescence (EL) dominant wavelength becomes smaller with current increasing. These improvements should be attributed to the reduced interface roughness of MQW and more uniformity of indium distribution in MQWs by the interruptions with TMIn-treatment.

Keywords: TMIn-treatment, InGaN/GaN quantum well, green LED

PACS: 78.66.Fd, 78.67.De, 85.60.Jb **DOI:** 10.1088/1674-1056/25/10/107803

1. Introduction

The quantum efficiency (QE) of InGaN-based light emitting diode (LED) at a wavelength in a range between 365 nm and 450 nm has been extensively studied and greatly improved recently. Blue LED can achieve external quantum efficiency of more than 70%.[1,2] However, InGaN-based LED suffers two critical issues, namely, efficiency droop with increasing operating current[3] and decrease of efficiency with increasing emission wavelength.[4] The QE of InGaN green LED is still relatively poor.[5] Several mechanisms have been proposed to explain this phenomenon. The large lattice mismatch between InN and GaN leads to low miscibility,[6] indium aggregation and phase separation usually occurring through spinodal decomposition.[7] Strain-induced polarization effect causes strong quantum confinement stark effect (QCSE), which reduces the spatial overlap of electron–hole wave function in the quantum well (QW), and therefore the radiative recombination efficiency of carriers.[8] Inefficient ammonia decomposition at the low growth temperature of multiple quantum wells (MQWs) results in indium (In) atoms accumulating on the surface, which forms defects, such as metallic In-clusters, nitrogen (N) vacancy and v-shaped defects.[9–11] The existence of defects degrades the crystalline quality and luminous efficiency of LED severely. For these reasons, tremendous effort has been made to improve optical properties of InGaN-based LED with green emission.

Growth interruption, as an efficient method, is often adopted to optimize the performances of InGaN LED. The effects of growth conditions are investigated systematically. Recent research showed that interruption in a hydrogen-free atmosphere is more favorable for obtaining green-spectral range LEDs.[12] The duration of interruption is crucial and it affects the surface morphology of QW and optical properties.[13–15] Moreover, trimethylindium (TMIn)-treatment in InGaN/GaN MQWs was also reported. Indium phase separation in MQWs was suppressed and the indium segregation was reduced, which was helpful in reducing the interface roughness of MQW.[16] Homogeneous indium composition leads to more monochromatic luminescence.[17–19] The output power of green LED could also be enhanced by TMIn-treatment.[20,21] Therefore, TMIn-treatment is used in growth process extensively to obtain green LEDs. However, the combination of metal organic chemical vapor deposition (MOCVD) growth interruption and TMIn-treatment has rarely been reported.

In our previous research, interruption in the growth pro-

*Project supported by the National Natural Science Foundation of China (Grant Nos. 11204360 and 61210014), the Science and Technology Planning Projects of Guangdong Province, China (Grant Nos. 2014B050505020, 2015B010114007, and 2014B090904045), the Research Fund for the Doctoral Program of Higher Education of China (Grant No. 20134407110008), the Guangzhou Municipal Science and Technology Project of Guangdong Province, China (Grant No. 2016201604030027), and the Zhongshan Science and Technology Project of Guangdong Province, China (Grant No. 2013B3FC0003).

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cess of MQWs was investigated.[22] A long wavelength (about 550 nm) LED, with smaller blue shift of dominant wavelength with current increasing is obtained. Based on this research, a new method of growing MQWs is provided to improve the performances of green LEDs. In contrast to the previous investigation, interruptions with TMIn-treatment are inserted repeatedly into the growth process of each QW. The effects of the multiple interruptions with TMIn-treatment and mechanisms are also discussed.

2. Sample structures and experimental procedures

Samples used in this study were grown on *c*-plane (0001) sapphire substrates. After substrates were thermally cleaned in hydrogen ambient for 6 min at 1010 °C and a 30-nm-thick GaN nucleation layer was grown at 530 °C, a 4-μm-thick n-GaN buffer with a doping concentration of 5×10^{18} cm^{-3} was followed at 1050 °C. Then five periods of InGaN/GaN MQWs with 20% nominal indium content were then deposited. Five interruptions with a duration of 18 s were adopted in each QW. During the interruption, Ga source was turned off, while TMIn, ammonia flow and other growth conditions remained unchanged.

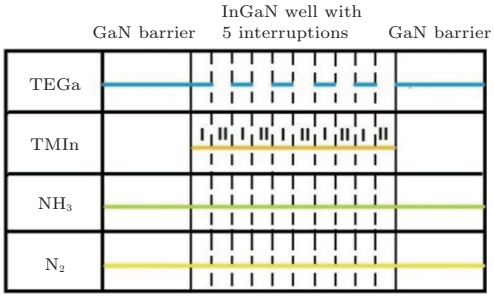


Fig. 1. (color online) Process of growing an InGaN/GaN period in LED A. Regions signed by I and II indicate the different growth conditions. I, TMIn collaborated with Triethylgallium (TEGa), II, growth interruption, TEGa was idle.

Figure 1 shows the growth conditions changing during the InGaN/GaN MQWs growth. Finally, a 200-nm-thick p-GaN with a doping concentration of 5×10^{18} cm^{-3} was grown. This sample was denoted as LED A. As a reference, another sample without interruptions in MQWs was also grown, and named LED B. For MQW interface characterization, the same structures as LED A and LED B without p-GaN layer were prepared and named MQW A and MQW B respectively. The LED chips, each with sizes of 300 μm×300 μm, were fabricated by photolithography and etching. Ni/Au and Cr/Pt/Au alloy were used for p- and n-type Ohmic contact, respectively. The structural properties of the two samples were investigated by a high-resolution x-ray diffraction (HRXRD) with a Panalytical system equipped with a four-bounce channel-cut Si (220) monochromator that delivered a pure CuKα_1 line of wavelength 0.154 nm. The interface roughness was investigated by grazing incident x-ray (GIXR) with the same system. Photoluminescence (PL) measurements were carried out with a 405-nm semiconductor continuous wave laser as an excitation source. The power of excitation source was 50 mW. Electroluminescence (EL) measurements from 1 mA to 100 mA current were also performed.

3. Results and discussion

HRXRD ω–2θ curves for the (0002) reflection of the InGaN/GaN MQWs for LEDs A and B are shown in Fig. 2(a). The vertical coordinate (intensity) for LED A is transformed in order to avoid overlap of the two curves in Fig. 2(a). The main peaks in two spectra originate from the n-GaN layer. The fittings of spectra indicate that the quantum well pairs of the LEDs have similar periodicities. Taking into account MQW growth conditions and results of PL measurements, the values of well thickness, barrier thickness and indium content are calculated to be 3 nm, 14.4 nm, 21% for LED A and 2.9 nm, 14.5 nm, 20.8% for LED B. The grazing incident x-ray (GIXR) 2Theta–Omega curves of MQWs A and B are shown in Fig. 2(b). The fittings of spectra show that quantum wells of the two MQWs have similar structures. However, the values of interface roughness are different: 1.3 nm for MQW A and 2 nm for MQW B. With growing the interruptions with TMIn-treatment, less phase separation and indium aggregation lead to smoother interfaces between quantum wells and barriers.

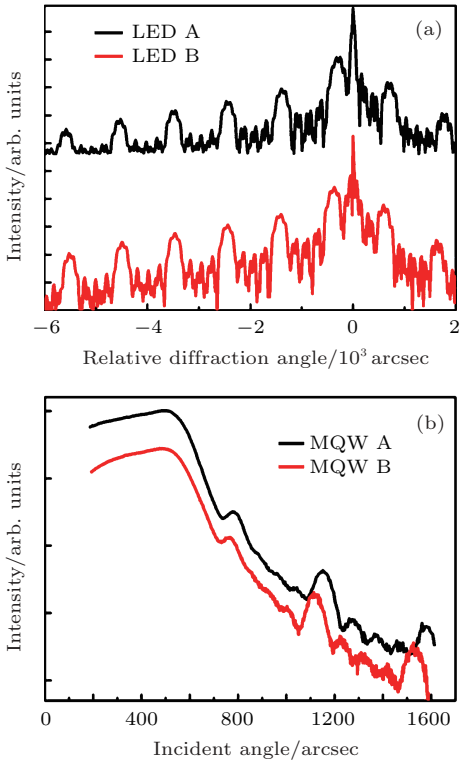


Fig. 2. (color online) (a) HRXRD curves for the (0002) reflection of the InGaN/GaN MQWs for LEDs A and B. The HRXRD periodicities of two LEDs were similar; (b) GIXR curves of the MQWs A and B. The surface roughness values of two samples are different.

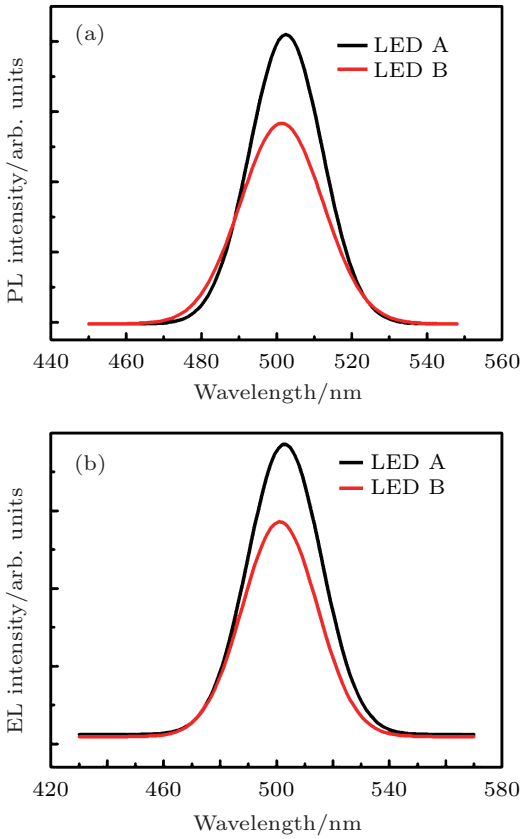


Fig. 3. (color online) (a) PL spectra of LEDs A and B at 300 K. The FWHM values of LEDs A and B are 22 nm and 27 nm respectively. (b) EL spectra of LEDs A and B at 20 mA. The integrated EL intensity of LED A is about 1.4 times higher than that of LED B.

Room-temperature PL spectra are measured to investigate the optical properties of two LEDs which are shown in Fig. 3(a). The PL intensity of LED A is higher than that of LED B. It indicates that the crystal quality of MQW in LED A is better than that in LED B. The dominant peaks of both LEDs show nearly no differences between 503 nm of LED A and 502 nm of LED B. However, the 22-nm full-width at half maximum (FWHM) of LED A is smaller than 27 nm of LED B. The EL spectra of LEDs A and B at 20 mA shown in Fig. 3(b) represent that the integrated EL intensity of LED A is much higher (1.4 times) than that of LED B. The light output powers (LOPs) at 20 mA of LED chips A and B are 3.13 mW and 1.91 mW respectively. The LOPs of both LEDs are not so high mainly because there are only 5 pairs of MQWs on LEDs and LEDs chips are not packaged. The EL FWHM of LED A is also narrower than that of LED B. PL and EL measurements indicate that the optical property and power of LED obtained by our growth method are improved. The reason for this improvement of LED A in Fig. 3 is the compositional uniformity and lower density of defects of indium. The indium content increases in the growing direction due to indium surface segregation.[23] The TMIn-treatment at initial interruptions suppresses the segregation. In addition, the TMIn-treatment smooths the growing surface because indium atoms are surfactant, which makes a more uniform indium distribution and smooth interface, as fitted from Fig. 2(b). Moreover, the TMIn-treatment suppresses InGaN decomposition to improve the efficiency of indium incorporation and interruptions suppress the indium aggregation to achieve a more homogeneous indium composition, a lower density of defects. Consequently, interruptions with TMIn-treatment reduce the FWHM of LED spectrum and improve the intensity of LED.

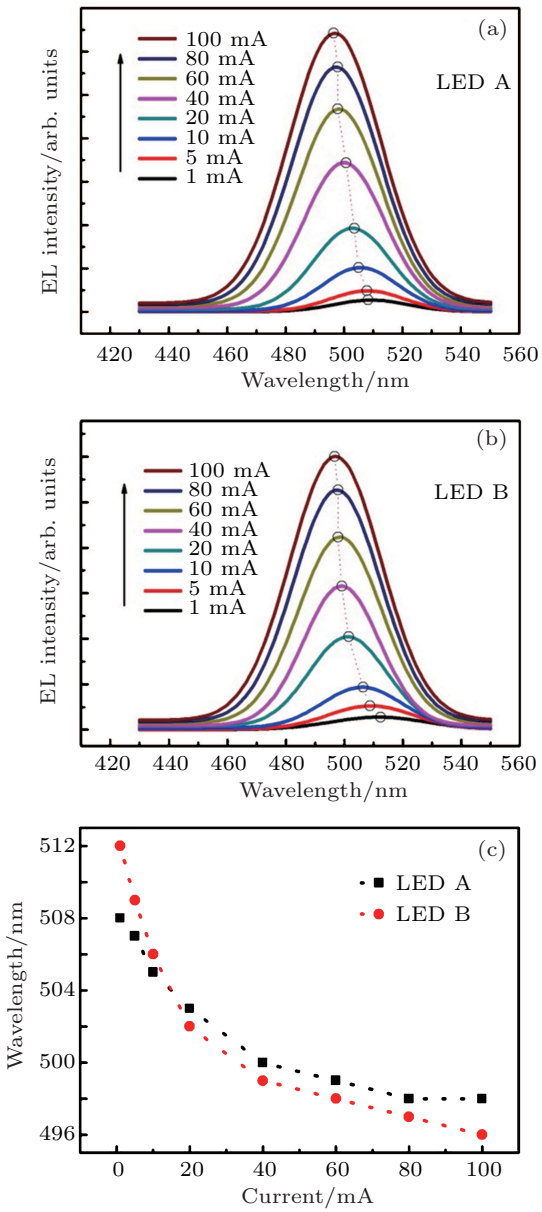


Fig. 4. (color online) (a) and (b) EL spectra of LEDs A and B at different amounts of current. The dominant peak positions are indicated by hollow circles. Dashed lines are guided for eyes; (c) the dominant peak positions of LED A and LED B at different amounts of current.

Another interesting problem of green LED is the stability of wavelength varying with injection current. To investigate the variation of wavelength with injection current, EL spectra of LEDs A and B at different amounts of current are given in Figs. 4(a) and 4(b). With current increasing from 1 mA to 100 mA, emission intensity increases. However, the blue-shifts of two LED chips are obviously different. As shown in

Fig. 4(c), blue shifts of LEDs A and B are 10 nm (from 508 nm to 498 nm) and 16 nm (512 nm to 496 nm) when current increased from 1 mA to 100 mA, respectively. The amount of blue shift of LED A is much smaller than that of LED B. The reason for this phenomenon is that the distribution of indium composition in QW becomes more uniform and localized energy states therefore decrease. Indium composition fluctuation in QW can be suppressed by interruptions with TMIn-treatment.

From the above discussions, it can be concluded that the superior performances of LEDs derive from the improvement of crystal quality, decrease of interface roughness and uniform indium composition of MQWs. In the process of TMIn-treatment, indium acts as surfactant which enhances the surface migration of nitrogen atoms.[24] Crystal quality and interface roughness of MQW will be improved due to improving surface migration of nitrogen atoms. In the process of interruptions, TMIn-treatment is proved to prevent indium segregation [25,26] and the indium composition becomes more uniform on MQWs.

4. Conclusions

In this work, we investigate the effects of multiple interruptions with TMIn-treatment indium in the process of InGaN/GaN MWQs. The interruptions and TMIn-treatment improve the performance of green LED collaboratively. More homogeneous indium composition and lower defect density lead to stronger but narrower luminescence, and few localized energy states lead to smaller blue shift as current increases. The research results provide important information for optimizing the performances of green LEDs.

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