Quasi-homoepitaxial GaN-based blue light emitting diode on thick GaN template*

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The high power GaN-based blue light emitting diode (LED) on an 80-μm-thick GaN template is proposed and even realized by several technical methods like metal organic chemical vapor deposition (MOCVD), hydride vapor-phase epitaxial (HVPE), and laser lift-off (LLO). Its advantages are demonstrated from material quality and chip processing. It is investigated by high resolution X-ray diffraction (XRD), high resolution transmission electron microscope (HRTEM), Rutherford back-scattering (RBS), photoluminescence, current-voltage and light output-current measurements. The width of (0002) reflection in XRD rocking curve, which reaches 173″ for the thick GaN template LED, is less than that for the conventional one, which reaches 258″. The HRTEM images show that the multiple quantum wells (MQWs) in 80-μm-thick GaN template LED have a generally higher crystal quality. The light output at 350 mA from the thick GaN template LED is doubled compared to traditional LEDs and the forward bias is also substantially reduced. The high performance of 80-μm-thick GaN template LED depends on the high crystal quality. However, although the intensity of MQWs emission in PL spectra is doubled, both the wavelength and the width of the emission from thick GaN template LED are increased. This is due to the strain relaxation on the surface of 80-μm-thick GaN template, which changes the strain in InGaN QWs and leads to InGaN phase separation.

Keywords: homoepitaxy, strain relaxation, metal organic chemical vapor deposition (MOCVD), hydride vapor-phase epitaxy (HVPE)

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1. Introduction

Various advantages of the freestanding GaN substrates to homoepitaxy of high power or ultraviolet (UV) light emitting devices have been reported by many researchers.[1–5] The 2 inch-diameter GaN substrates are mainly fabricated by hydride vapor epitaxy (HVPE) due to the high growth rate at 100 μm/h and low threading dislocation density (TDD) of 10^6 cm^{-2}–10^7 cm^{-2}. The decrease of dislocations will reduce non-radiative recombination and increase the thermal conductivity of the GaN epilayers.[6] The GaN substrates are not only lattice and thermal matched to GaN epilayers, but also provide a strain-free surface for InGaN active layer to grow.[7] Gorgen *et al.* proposed that a fully relaxed sample is able to incorporate more indium during its process of growth than a strained sample.[8] This means that a higher quality and higher In content InGaN active layer can be grown on GaN substrate at a higher temperature than that on a strained surface. Furthermore, the GaN substrate has a higher thermal-conductivity than sapphire and it can avoid the phonon mismatch and the thermal interface barrier.[4] GaN-based LED grown on GaN substrate may be operated under an order higher injection level than a conventional one.

However, the commercial GaN-based LEDs are seldom fabricated directly using the GaN substrate, because its cost is two orders higher than that of sapphire substrate. It is reasonable to use the thick GaN template on sapphire to grow GaN-based LED structures since the properties of the thick GaN template are similar to those of the GaN substrate. The high quality GaN can be obtained by HVPE when the film thickness reaches 80 μm. Speck *et al.* found that the TDD in relatively thick GaN layers is inversely proportional to the film thickness.[9] The TDD of 80-μm-thick GaN template corresponds to 10^7 cm^{-2}, which is two orders lower than that of several-micron-thin GaN film on sapphire substrate. According to Reynolds' results, the surface is strain free at this thickness,[10] and indium is easily incorporated into the QWs due to the thermodynamic properties changing.[11] The growth on the strain-free surface also leads to less quantum confined Stark effect (QCSE) in multiple quantum wells (MQWs).[11] Moreover, in our earlier work,[12] it was found that the tilt of the crystal columns becomes smaller as the film thickness increases. The thick GaN template is not expensive because of the following three facts. The first is that the growth rate of GaN is high via HVPE technique. Secondly, thickness of the GaN template is much smaller than that of GaN substrate, and thirdly, the grinding and polishing are not needed for regrowth. In fact, the grinding and polishing processes not only increase

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the cost, but also are harmful to the expired GaN substrate.[13] Furthermore, the thick GaN template can act as a freestanding substrate after laser lift-off (LLO) from sapphire in chip processing, since the normal thickness of the commercial GaN-based LED chip is about 80 μm. It cuts down the cost substantially compared to chip processing on the GaN substrate.

In spite of the above advantages of a thick GaN template, there are not many works reported on the GaN-based LED on thick GaN template.[11,14–16] The GaN template is a virtual substrate since the real substrate underneath is sapphire. Hence, we call the regrowth on the GaN template "quasi homoepitaxy". Sakai *et al.* reported that the InGaN/GaN MQW grown on an 80-μm GaN template show PL intensity three times higher and lower reversal leakage than those on sapphire.[14] The results were attributed to the lower TDD in the epilayer, which causes the sharp interfaces at InGaN/GaN and GaN:Mg/GaN. They did not take care of the growth interface between HVPE-GaN and metal organic chemical vapor deposition (MOCVD)-GaN. Johnson *et al.* have studied the effect of GaN template layer strain on the growth of InGaN/GaN MQW LED.[11] They found that the thermodynamic limitation of different template growth surfaces is changed by strain relaxation, which leads to more In incorporation on the thicker template. They also concluded that the residual strain in the template can strain the entire devices, especially in the active region. In contrast to the HVPE GaN template, when their templates were grown by MOCVD, a thicker template had a dislocation density about five times the dislocation density of a thinner one. Recently, some researchers have reported that InGaN/GaN MQW LED had grown on about 30-μm-thick HVPE GaN template.[15,16] Both of them observed the improvements of crystal quality and luminous efficiency of the LEDs on thick GaN template. Sun *et al.* also showed that the LLO process had less degraded effect on the LED on thick template than that on a conventional buffer layer.[15] Actually, the growth on the thick GaN template is reported much less than that on the GaN substrate. Besides, growth surface treatment to eliminate the interface between HVPE-GaN and MOCVD-GaN[17,18] and optimization of growth condition to inherit the low TDD and residual strain of HVPE-GaN[19,20] for GaN substrates are also essential to regrowth on the thick GaN template.

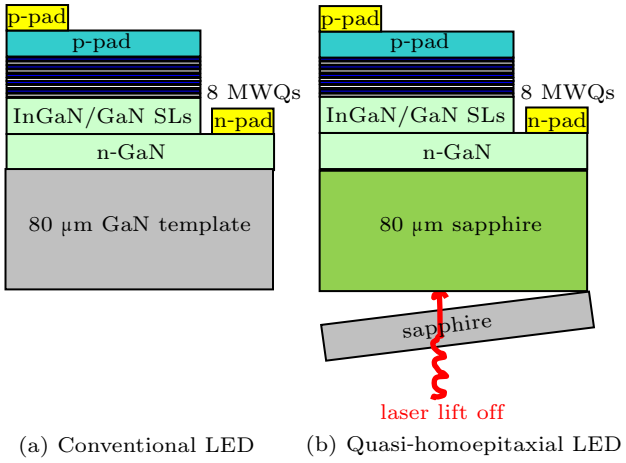
In this work, 80-μm-thick GaN templates are fabricated on GaN/sapphire substrates using HVPE. Then, GaN-based LED is grown on the thick GaN template using MOCVD. The LED chips are fabricated by LLO and conventional processes. Then, we choose a template with 80-μm-thick GaN, because such a template costs less than a 400 μm–500 μm freestanding substrate and also makes it easy to control the concave bowing in the regrowth process. The quasi-homoepitaxial GaN-based LEDs are investigated by high resolution X-ray diffraction (HR-XRD), Rutherford back scattering (RBS), high resolution transmission electron microscopy (HRTEM), photoluminescence (PL), and electroluminescence (EL).

2. Experiment

Firstly, an undoped 4-μm-thick GaN/sapphire template (labeled as sample A) was grown on sapphire substrate, which is the most widely used substrate for GaN crystal growth, by MOCVD in a Thomas Swang reactor using the traditional two-step way. Secondly, 80-μm-thick GaN film (labeled as sample B) was grown on the 4-μm-thick GaN template by HVPE at 1090 °C in a conventional vertical reactor. The buffer layer was heated up to the HVPE process growth temperature in ammonia atmosphere in order to be protected from decomposition. It was unintentionally doped and had a carriers mobility of 193 cm^2/Vs. Thirdly, the blue LEDs were grown by MOCVD on the 4-μm-thick GaN template (labeled as sample C) and 80-μm-thick GaN template (labeled as sample D) in the same run for comparison. The LED structure consisted of a 2-μm undoped GaN layer, a 2-μm Si-doped GaN layer, eight periods of InGaN (2.5 nm)/GaN (12 nm) MQWs, and a 0.1-μm Mg-doped GaN layer. The samples C and D were processed into square mesas of 1 mm × 1 mm in size, by photolithography and inductively coupled plasma (ICP) reactive ion etching. The contacts of Cr/Pt/Au and ITO transparent current spreading layer were deposited to n- and p-GaN layers, respectively. For sample C, to obtain blue LED chips, it was reduced to 80-μm thickness by grinding and polishing and then was separated by the laser scriber and broker. For sample D, it was bonded to a glass and subjected to the LLO process. Subsequently, the glass carrier was removed by acetone, and the 80-μm-thick free-standing film LED chips were obtained successfully. Figure 1 shows the structure of samples C and D. Instead of grinding and polishing, it just needs LLO. What is more, since the material in n side is GaN, it is possible to make vertical structure LED, which would show higher performance. Finally, the chips were packaged with silicone resin for subsequent measurements.

The XRD rocking curves (ω scans) and 2θ–ω scan were measured using a Philips materials research diffractometer equipped with a four crystal monochromator, delivering a pure Cu K_α 1 line of wavelength, $\lambda = 0.15406$ nm. The HRTEM images displayed the detailed parameter of MQWs and defect distribution. The accurate In component was deduced by RBS measurement. The strain in InGaN QWs was determined by using the lattice parameters, In component following the procedure described in detail in Refs. [21]–[22]. PL spectra were measured with a Renishaw via Raman microscope at room temperature, and the excitation wavelength of laser is 405 nm. EL spectra were obtained by a CCD fiber spectrometer. For measurement of their voltage versus current (I–V)

curves, the LED chip was operated in pulsed current-injection mode to eliminate Joule heating, while the light output versus current (L–I) curves were measured under continuous wave (cw) mode to show the different performance of heat dissipation. The light output of the devices was measured using an integrating sphere.



(a) Conventional LED (b) Quasi-homoepitaxial LED

Fig. 1. (color online) Structures of (a) conventional and (b) quasi-homoepitaxial LED.

3. Results and discussion

The crystalline quality of 2-inch GaN-based wafers on sapphire and thick GaN template, before and after LEDs growth was estimated by HRXRD rocking curve measurements. Figure 2 shows the rocking curves of XRD along (0002) reflection for sample A (solid black square), sample B (solid red circle), sample C (hollow green triangle), and sample D (hollow blue diamond), respectively. Their full widths at half maximum (FWHMs) are 254″, 130″, 258″, and 173″, respectively, as shown in Table 1. For sample D growth process, the FWHM decreases from 254″, to 130″ and then increases to 173″, while for sample C, it changes from 254″ to 258″. It indicates that the crystal quality of 80-μm-thick GaN template is greatly increased after HVPE re-growth. However, it decreases slightly after LED structure re-growth by MOCVD. However, the crystal quality of sample D is much better than that of sample C. This high crystal quality would lead to the high performance of the 80-μm-thick free-standing LED.

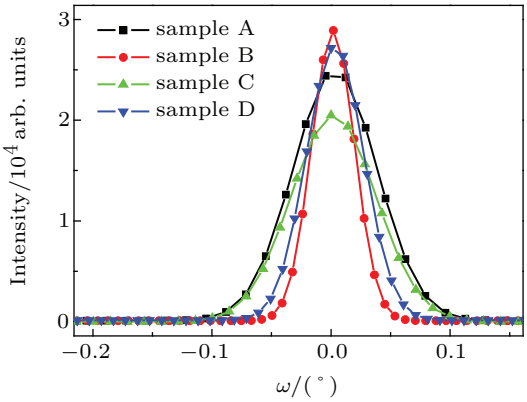


Fig. 2. (color online) Rocking curves of XRD along (0002) reflection for samples A, B, C, and D.

The HRTEM images of the quantum wells of the conventional and freestanding structure LED are shown in Fig. 3. The thicknesses of quantum well (d_w) and quantum barrier (d_b) are obtained by using the software digital micrograph, as shown in Table 2. The detail of the TEM images show that the quantum wells in sample D have a sharper interface and less defects than those in sample C. It means the thick GaN template leads to a high crystal quality of the LED wafer, which is consistent with the XRD results. On the other hand, the period of sample D is a little more than that of sample C, which confirms that the growth rate is increased by thermodynamic limitation changes.[11]

Table 1. Information and XRD results of samples A, B, C, and D.

Sample	A	B	C	D
Sample information	4-μm-thick GaN/sapphire template grown by MOCVD	80-μm-thick GaN film grown by HVPE	Blue LED on sample A	Blue LED on sample B
(0002)FWHM/(″)	254	130	258	173

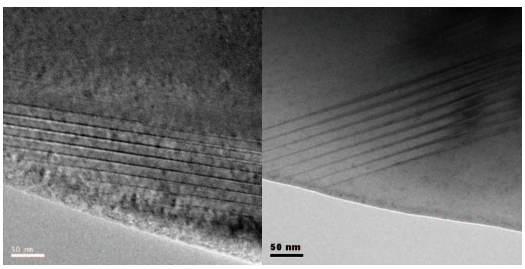


Fig. 3. HRTEM images of (a) sample C and (b) sample D. From the top to the bottom, the layers are n-GaN, electron reservoir layer, MQW, and p-GaN, respectively. Sample D image shows the sharp interfaces and low defects in MQW.

Table 2. Structure parameter of MQW.

Sample	d_b/nm	d_w/nm	Period/nm	Period number
Sample C	13.12	2.49	15.61	8
Sample D	13.43	2.50	15.93	8

Figure 4 shows the random RBS spectra of the two LEDs. They are almost the same, which means that they have the same composition. The average indium compositions in the pairs of InGaN/GaN layers for both samples are 0.031. As the parameter of quantum wells shows in Table 2, the In composition is 0.194 for sample C and 0.198 for sample D.

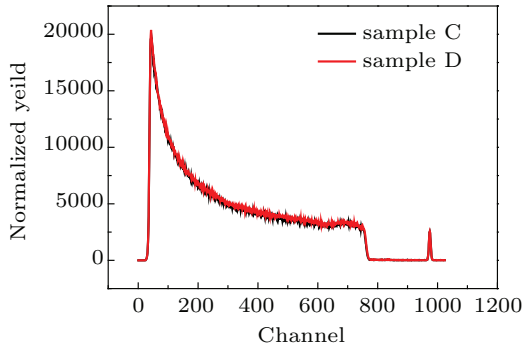


Fig. 4. (color online) Random RBS spectra of sample C and sample D.

Figure 5 shows the double-crystal XRD (ω–2θ) curves of samples C and D. From these XRD curves, the -3 to $+3$ order satellite peaks are clearly observed. Besides, the average distances between two satellite peaks of samples C and D are similar as $2160''$ by which the quantum well period can be calculated to be 15.4 nm as designed. The positions of GaN (0002) of samples C and D are 34.5390 and 34.5590, respectively as shown in the inset of Fig. 5. The angular separation between the $m = 0$ fringe of InGaN/GaN quantum well structure and the GaN peaks for samples C and D are 0.1450 and 0.1610, respectively. These distinct satellite peaks indicated that the interface of the well and barrier is smooth and sharp. Upon the similarity of the two profiles of the diffraction curves, it can be concluded that the widths of the quantum wells and barriers in both samples are almost identical for samples C and D. The differences of the GaN peak position indicated that the strain for sample D is lower than that of sample C.

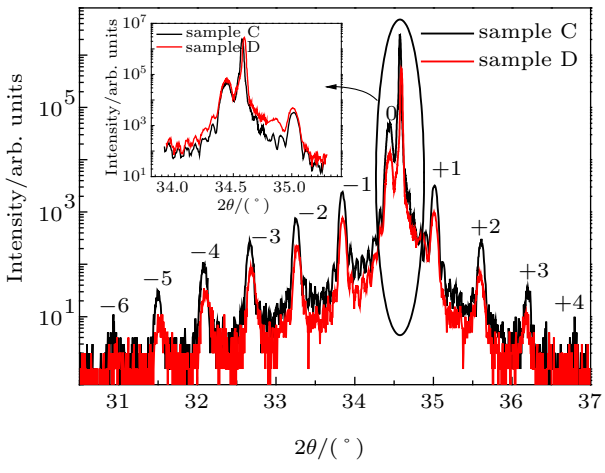


Fig. 5. (color online) The (ω–2θ) XRD curves for samples C and D.

We simulate the XRD results by using the parameters, which are obtained from the TEM, RBS, and XRD measurements, as shown in Fig. 6. Based on these indium compositions, thickness of quantum wells (QWs) and quantum barriers (QBs), the symmetric and asymmetric reflection curves of XRD, the strain of InGaN wells can be extracted as follows.

The perpendicular strain can be calculated as $+0.17\%$ and $+0.15\%$ for samples C and D, respectively. According to the d spacing formula of (1012) plane for wurtzite and the c value determined from the (0002) diffraction, we obtain the experimental result and the parallel strain can be calculated as -0.273% and -0.107% for samples C and D, respectively. Consuming the strain in GaN barriers is identical to GaN substrate and the residual strain in GaN substrate is zero, the parallel strains in InGaN QWs are calculated as -1.71% and -0.789% for samples C and D, respectively. If the InGaN QWs are fully strained to GaN barriers, the strains are 2.13% and 2.18% for samples C and D, respectively. Considering the parallel strain in GaN substrate of sample C is compressive, the parallel strains in InGaN QWs are nearly the same as the fully strained value. However, the parallel strain in 80-μm-thick GaN template of sample D is fully relaxed or a bit tensile according to Refs. [8], [10], and [11]. The parallel strain in InGaN QWs of sample D is higher than -0.789%. It is indicated that the strain in InGaN QWs grown on the thick GaN template is relaxed about 64% for sample D. According to the above results, the LED structure grown on GaN thick template shows higher crystalline quality, similar In incorporation into InGaN QWs and most relaxed QWs.

Figure 7 shows the room temperature PL spectra for samples C and D, respectively. The excitation wavelength is 405 nm. The presence of Fabry–Pérot interference fringes for sample C is a characteristic feature for LEDs grown on sapphire as expected. They are absent in sample D due to the weak interference caused by the 80-μm-thick GaN film. The MQWs emission from sample D is about 30% stronger than those of sample C and the FWHMs of PL spectra are 29 nm and 18 nm for samples C and D, respectively. According to the above results of XRD and TEM measurements, the high crystalline quality and strain relaxation in the InGaN QWs in sample D increase the radiative recombination rate in QWs and shorten the emission bandwidth. However, the emission peak is located at 468 nm for sample D, which is 10-nm redshift compared to that for sample C. It indicates that InGaN phase separation seems to have occurred. InGaN phase separation may arise in strain-relaxed InGaN because of the high miscibility gap between InN and GaN.[23] It is proposed that there is fully relaxed growth surface on the 80-μm GaN template.[8] Since the strain-relaxed growth surface may lead to higher quality InGaN QWs and weaker indium fluctuation, some driving forces about the thermodynamics factors may appear in the following growth of barriers, e-blocking layer or p-GaN layer, which will lead to the InGaN phase separation. Wang *et al.* thought that the appropriate degree of phase separation might also enhance the localization of carriers and light emission.[24]

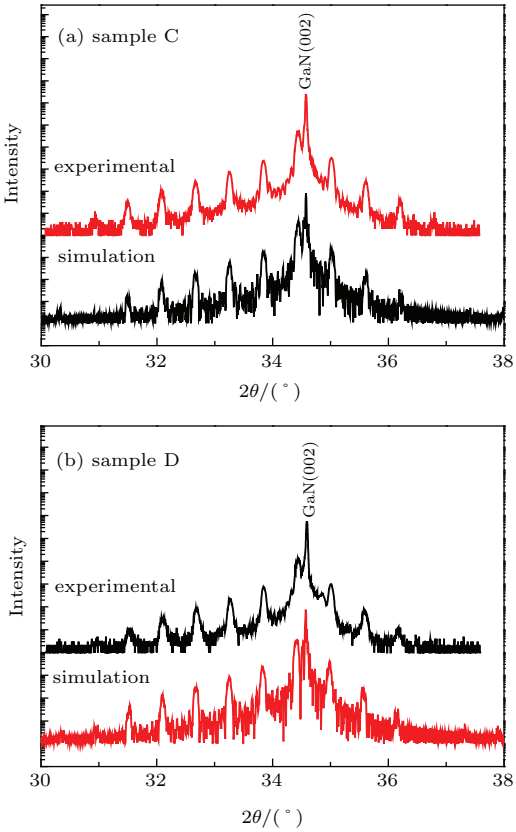


Fig. 6. (color online) Simulation and experimental HR-XRD curves of sample C (a) sample D (b).

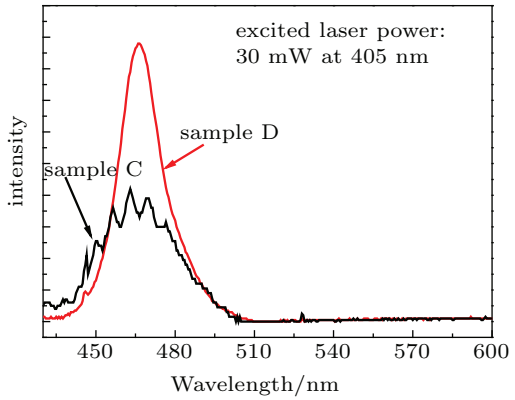


Fig. 7. (color online) Room-temperature PL spectra of samples C and D. The excitation wavelength is 405 nm.

The forward voltage for the sample D at 350 mA is 3.40 V, which is a little smaller than 3.47 V for sample C. This difference in the forward voltage could be attributed to the three following reasons. (i) The low body resistance of n-type GaN for sample D due to its 80-μm thickness; (ii) The smaller compression strain of sample D leads to the weaker polarization which will reduce the height of the triangular barrier and thus the forward voltage; (iii) The low series resistance of p-type GaN for sample D due to its high quality p-type GaN.[14] What is more, when the injection current level was higher, the difference of voltage was larger. This indicates that it has more advantages for the LED performance under the higher injection level.

Figure 8 shows *L–I* curves of samples C and D and the inset shows the normalized EL spectra with 451-nm and 464-nm emission wavelengths for samples C and D, respectively. The intensity of the light output is higher for sample D than that for sample C, especially under high injection current level. The intensity of the light output at 350 mA of sample D is almost twice that of sample C. Besides, the saturated injection current of samples C and D is 900 mA and 1050 mA, respectively. The light output enhancement for sample D is due to the high crystalline quality and large strain relaxation in InGaN QWs, which is similar to PL results. Moreover, the high thermal conductivity of GaN substrate also plays an important role in the enhancement for sample D after removing the sapphire substrate. There are less phonon mismatches and the thermal interface barrier, and there are less nonradiative recombination centers where the heat is generated for sample D. This good heat dissipation also leads to high saturated injection current level; on the other hand, it is found that the emission wavelength is smaller than that in PL result because of the carrier screening effect. Emission from the sample C shows 7-nm blueshift, while that for sample D is about 4 nm. It is due to the strain relaxation in QWs of about 64% for sample D. The large relaxation will weaken the effects of carrier screening and laser lift-off on emission blue-shift for sample D. It indicates that the emission wavelength for thick GaN template LED is more stable with the current increasing. Incidentally, the light output should be enhanced further when the thick GaN template LED is fabricated into a vertical structure.

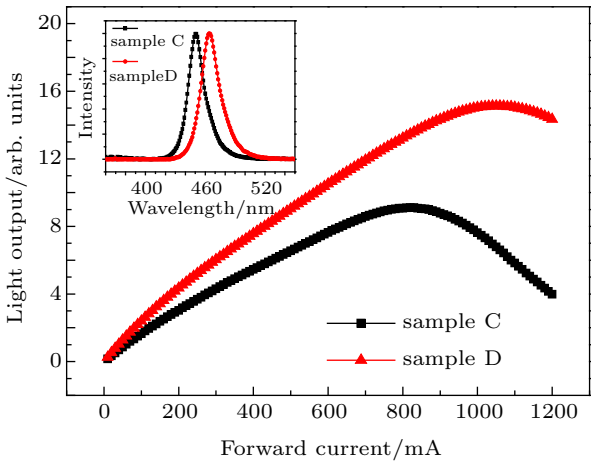


Fig. 8. (color online) The *L–I* curves under cw measurement mode of samples C and D. The inset shows the normalized EL spectra.

4. Conclusion

In summary, GaN-based blue LED on 80-μm-thick GaN template with emission wavelength of 464 nm was fabricated successfully by MOCVD, HVPE and LLO techniques. According to the results of the XRD curves, TEM images, RBS

analysis, PL spectra, *I–V* and *L–I* curves, it shows better structural, including crystal quality and strain relaxation, electrical and optical properties than those of the reference one. On the other hand, comparing to the reference blue LED, wavelength shift was observed in this thick GaN template LED device. It was mainly attributed to the relaxed strain in the 80-μm-thick GaN. Anyway, by using the quasi-homoepitaxy growth, thick GaN template LED shows much potential for the application for high power, high injection current level and high efficiency LEDs and laser diodes.

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