In this paper, we report the fabrication of an In$_{0.2}$Ga$_{0.8}$N/GaN multiple-quantum-well (MQW) structure on vicinal sapphire substrates with a very small offset angle of 0–1° by low-pressure metallicorganic chemical vapor deposition. Our study demonstrates that the quality of the In$_{0.2}$Ga$_{0.8}$N/GaN MQW structure is very sensitive to the offset angle of the vicinal substrate. High-resolution X-ray diffraction analyses demonstrated high-order satellite peaks and clear fringes between them for all MQW structures fabricated, from which the interface roughness (IRW) was estimated. The IRW of the In$_{0.2}$Ga$_{0.8}$N/GaN MQW structure fabricated on 0.2° off sapphire substrate was determined as 1.36% of the quantum well layer period. Besides, reciprocal lattice mapping was employed to examine the strain status of the MQW. The lattice relaxation of the same specimen mentioned above was estimated to be $7.4 \times 10^{-5}$. It is therefore manifested that an In$_{0.2}$Ga$_{0.8}$N/GaN MQW structure with abrupt interfaces and good layer periodicity was grown. From it, a shortest radiative lifetime of 14.2 ns and a lowest fluctuation of 5.6 meV in the emission energy of micro-photoluminescence mapping were achieved. In addition, superior material qualities of the whole film fabricated on 0.2° off substrate were recognized by cross-sectional transmission electron microscopy. Based on the results mentioned above, a high-quality In$_{0.2}$Ga$_{0.8}$N/GaN MQW blue light, emitting diode (LED) has been fabricated on 0.2° off substrate, which demonstrated a strong room-temperature electroluminescence emission at the wavelength of 465 nm and with the full width at half maximum of only 19 nm. The same device also showed an output power of 13.4 mW and an external quantum efficiency of 19.2%. Both these characteristics are improved drastically compared with the devices fabricated on the substrates with other offset angles. Conclusively, the use of an appropriately misoriented sapphire substrate is suggested to be effective for elevating the emission efficiency of In$_{0.2}$Ga$_{0.8}$N/GaN MQW blue LED fabricated thereon.

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n-GaN epilayer, a five-period In0.02Ga0.98N/GaN prestained layer, a five-period In0.2Ga0.8N/GaN active layer, a 0.1 μm thick p-AlGaN window layer, and a 0.1 μm thick p'-GaN contact layer. The thickness of the In0.2Ga0.8N well and GaN barrier in the active layer was around 3.1 and 12.1 nm, respectively, while the thickness of the prestrained layer was about 2.8 nm. In addition, all the samples were grown in the same run and therefore the growth conditions were completely the same except for the substrate misorientation. Hence, any variances in the crystalline quality of the top layers are reasonably considered to result from the substrate misorientation.

After epitaxial growth, the top ohmic contact to In0.2Ga0.8N/GaN MQW LEDs was formed using a metal system of Ni/Al/Au for the p'-GaN contact layer. Otherwise, the Ti/Al system was evaporated onto the exposed n-GaN epilayer. Finally, In0.2Ga0.8N/GaN MQW LEDs were cut into square pieces with a dimension of 300 × 300 μm.

The crystalline quality of our epitaxial structures was evaluated by high-resolution X-ray diffraction (HRXRD) using Cu Kα radiation as the X-ray source, and the corresponding reciprocal space mapping (RSM) was used to investigate the strain status of MQWs fabricated. Optical properties were analyzed spatially and energetically by micro-photoluminescence (μ-PL) mapping measurements performed at room temperature. The μ-PL spectra were excited with the 325 nm line of a He–Cd laser at an excitation power of 20 mW. Besides, the radiative lifetime was determined from the time-resolved PL (TRPL) measurements conducted at room temperature. In this case, the signal was excited by a GaN semiconductor laser (λ = 396 nm) with a pulse width of about 0.15 ns and a power density of 0.2 W/cm². Moreover, the distribution and the threading behaviors of dislocations in the specimens were studied by bright-field transmission electron microscopy (TEM). Finally, the electroluminescence (EL), current–voltage (I–V), and light–current characteristics of In0.2Ga0.8N/GaN MQW LEDs were measured at room temperature under a forward voltage of 3.5 V.

Results and Discussion

Figure 2 shows the HRXRD ω/2θ scan for the In0.2Ga0.8N/GaN MQW LED epitaxial structures fabricated on the sapphire substrates with an offset angle of (a) 0°, (b) 0.2°, (c) 0.35°, and (d) 1°, which are henceforth denoted as samples A, B, C, and D, respectively. As can be seen, all the HRXRD patterns demonstrate periodical structures, which can be attributed to the In0.2Ga0.8N/GaN MQWs fabricated. The strongest peak is due to the GaN layer, and all spectra clearly show high-order MQW diffraction peaks with the third-order satellite peak still being observable, indicating good layer periodicity. Besides, the MQW period can be determined from the positions of the MQW satellite peaks. It suggests that the grown structure also has abrupt interfaces between GaN barrier and InGaN well. The interface roughness (IRN) of MQW structures fabricated on various substrates was further analyzed by using the following equation:

$$W_n = W_0 + (ln \frac{2}{\Lambda}) \frac{n \Delta Q_x \sigma}{\Lambda}$$  [1]

where \(n\) is the order of the satellite; \(\Lambda\) and \(\sigma / \Lambda\) are the periods of satellite peak and IRN, respectively; \(\Delta Q_x\) is the angle distance between adjacent satellite peaks; \(W_0\) and \(W_n\) are the full-width at half-maximum (fwhm) of the zeroth- and n-th-order peaks, respectively. The inset of Fig. 2 shows the variation of MQW IRN as a function of substrate offset angle. As can be seen, the IRN of InGaN/GaN MQWs reaches a minimum of about 1.35% for the MQW structure grown on 0.2° offset substrate (sample B). As indicated in Ref. 9, the IRN of MQWs is affected by the defects, microstructure, and phase separation in MQWs. Therefore, our X-ray analysis results manifest that the crystalline quality of the MQW epitaxial structure fabricated on the substrate offset by 0.2° is superior to those fabricated on other substrates.

The strain status of the MQW systems fabricated was evaluated with the help of RSM around the asymmetric (1 0 1 5) Bragg reflections. Figure 3 shows the RSM patterns for samples A, B, C, and D, where the diffraction intensities are plotted in contours. Compared with samples A, C, and D, the RSM intensity from sample B is gathered. In general, the spread of diffraction intensity is related to the orientation distribution and decrease in the coherency of the scattering along the structure.10 This means that the GaN epilayer grown on the substrate with an offset angle of 0.2° has a better crystalline quality. Moreover, the reciprocal lattice point of satellite peaks up to the fourth order can be clearly observed for sample B, while the reciprocal lattice point of satellite peaks became relatively weak for samples A and C and almost vanished for sample D, suggesting that the superior quality of the GaN epilayer could be responsible for the achievement of a high-quality InGaN/GaN MQW structure on top. One can understand the overall strain state of the MQW structure with respect to the GaN epilayer by RSM results. By using the x positions of the reciprocal lattice points of GaN and MQW satellite peaks, namely \(Q_x^{GaN}\) and \(Q_x^{InGaN}\), in Fig. 3, the degree of lattice relaxation for an MQW structure, \(e_{\text{LS}}\), can be evaluated by the following equation:

$$e_{\text{LS}} = \frac{Q_x^{GaN}}{Q_x^{InGaN}} - 1$$  [2]

Hence, the degree of lattice relaxation for samples A, B, C, and D was evaluated to be about $1.28 \times 10^{-2}$, $7.4 \times 10^{-3}$, $8 \times 10^{-3}$, and $3.64 \times 10^{-4}$, respectively, with a deviation value lower than 10%. Evidently, sample B has the smallest degree of lattice relaxation. For a heteroepitaxial structure, the density of defects, such as threading dislocations, V defects, and so on in MQWs will increase with an increase in the lattice relaxation degree of the MQW structure. In other words, the InGaN/GaN MQW epitaxial structure of sample B seems to exhibit the lowest defect density compared with other samples. Hence, the results from the RSM analyses are in good agreement with those presented by the theoretical studies of IRN described above.

Figure 4 presents the TRPL data of samples A, B, C, and D obtained at room temperature at the energy of 2.76 eV (450 nm). Notably, the normalized intensity is plotted on a semilogarithmic scale vs time on a linear scale. The transients can be well fitted by the stretched exponential, and the fitting equation is given below:

$$I(t) = I(0)\exp \left[ - \left( \frac{t}{\tau} \right)^{\beta} \right]$$  [3]

where \(I(0)\) and \(I(t)\) are the PL intensity at time 0 and \(t\), respectively; \(\tau\) is the lifetime of the carrier. The stretching parameter \(\beta\) varies...
between zero and unity. Using Eq. 3, the carrier lifetime of samples A, B, C, and D extracted is about 18.4, 14.2, 17.9, and 25.4 ns, respectively, which is displayed in the inset of Fig. 4. The influence of the offset angle of the substrate on the carrier lifetime of InGaN/GaN MQWs is obvious. Namely, a lowest carrier lifetime for the InGaN/GaN MQW structure was achieved when the 0.2° off substrate was used. Moreover, Fig. 5a–d displays the micro-PL mapping images of samples A, B, C, and D, respectively, within a scanning area of 20 × 20 μm. Here, the fluctuation of emission energy over the area scanned is denoted as ΔE (in meV). As can be seen, the ΔE of samples A, B, C, and D is estimated to be about 25, 5.6, 11, and 71.2 meV, respectively. It is obvious that, compared with other samples, sample B shows a quite uniform emission spectra over the region scanned, where the fluctuation of emission energy is only 5.6 meV. Both the results demonstrated in Fig. 4 and 5 can be understood as follows. In general, the quantum-dot-like structures in InGaN/GaN MQWs could be responsible for the surprisingly high quantum efficiencies. Nevertheless, the nonuniform distribution of indium and phase separation of InGaN will procure the formation of microstructures in MQWs, which induces the localization of carriers within the MQW stack, and therefore lengthens the carrier lifetime. However, a lower fluctuation of emission energy.
Figure 6. TEM cross-sectional images of In_{0.2}Ga_{0.8}N MQWs fabricated on (a) 0° off, (b) 0.2° off, and (c) 1° off sapphire substrates. The diffraction condition is {0 0 2}.

Figure 6a-c show the bright-field TEM cross-sectional images of In_{0.2}Ga_{0.8}N/GaN MQWs grown on sapphire substrates with various misorientations. As can be seen from Fig. 6a, the GaN epilayer grown on 0° off substrate has many dislocations threading through the MQWs to the top surface of the epilayer. The TDD for this sample is estimated to be 1.6 \times 10^9 \text{ cm}^{-2} at the bottom of the n-GaN layer while it reduces to 6.3 \times 10^8 \text{ cm}^{-2} at the top of the n-GaN layer and 2.2 \times 10^9 \text{ cm}^{-2} in the p-GaN region. Otherwise, for the epilayer grown on 0.2° off substrate, many fewer dislocations are observable within the range in view. As shown in Fig. 6b, the TDD at the bottom of the n-GaN layer is about 9.4 \times 10^8 \text{ cm}^{-2}; however, the TDD at the top of the n-GaN layer reduces to 3.0 \times 10^8 \text{ cm}^{-2} and it is only 1.0 \times 10^8 \text{ cm}^{-2} in the p-GaN region. Finally, as demonstrated in Fig. 6c, quite a large number of dislocations are present in the whole film on 1° off substrate. Hence, for this specimen the TDD evaluated at the bottom of the n-GaN region is as high as 2.3 \times 10^9 \text{ cm}^{-2}; even at the top of this layer there is still a TDD of 1.2 \times 10^9 \text{ cm}^{-2} and also a TDD of 6.2 \times 10^8 \text{ cm}^{-2}

In the p-GaN region. For this degree of TDD a relatively rough top surface can be found as demonstrated in Fig. 6c.

Finally, the LED devices fabricated on the basis of the material structure of InGaN/GaN MQWs grown on the sapphire substrates with different offset angles were characterized. Figure 7a shows room-temperature EL spectra of In_{0.2}Ga_{0.8}N MQW LEDs fabricated on sapphire substrates with misorientations of 0, 0.2, 0.35, and 1°, respectively. Also, the inset of Fig. 7a shows the emission intensity and fwhm of EL emission plotted as a function of the substrate misorientation. Obviously, a strongest emission at 465 nm with a smallest fwhm of about 19 nm was achieved for the device fabricated on 0.2° off substrate. In addition, Fig. 7b gives the light output power as a function of the forward current for all the InGaN/GaN MQW blue LEDs fabricated. Generally, the commercialized LEDs operate at a forward current of 20 mA. However, as exhibited in the inset of Fig. 7b, the I-V characteristics of the LED fabricated on 0.2° off substrate (LED sample D) differ somehow from those of the LEDs fabricated on other misoriented substrates. Hence, in our case, the output power of LEDs was obtained as follows. For the LEDs fabricated on 0, 0.2, and 0.35° off substrates (LED samples A, B, and C, respectively), the output power was obtained under the same forward current but a different forward voltage of 3.7 V. Namely, for LED sample D the output power was obtained from an emission with the wavelength of 458 nm, while for LED samples A, B, and C the output power was obtained from an emission with the wavelength of 458 nm. Resultantly, the light output obtained from LED samples A, B, C, and D was 10.3, 13.4, 12.2, and 7.2 mW, respectively. Simultaneously, the external quantum efficiency of LED samples A, B, C, and D were estimated as 14.7, 19.2, 17.4, and 10.3%, respectively. All the output characteristics of LED sample B were superior to those of other LED samples, which help to further recognize the merit of using the 0.2° off substrate for the fabrication of InGaN/GaN MQW blue LEDs.

From the analyses described above, the possible mechanisms for employing the 0.2° off substrate to fabricate superior InGaN/GaN MQWs are considered as follows. Several past reports presented by Shen et al. indicate that 2° off sapphire substrates can be used to reduce the density of threading dislocations in GaN epitayers by about one order of magnitude and upgrade the performance of devices fabricated on them.\textsuperscript{18-21} These results were obtained from the epitaxial technique of plasma-assisted molecular beam epitaxy and a mechanism for the reduction of defect density proposed by Shen et
Conclusions

In summary, we have fabricated an $\text{In}_0.2\text{Ga}_{0.8}\text{N}/\text{GaN}$ MQW epitaxial structure on vicinal sapphire substrates by LP-MOCVD. X-ray measurements showed that the MQW structure fabricated on the 0.2° off substrate exhibited sharp interfaces between the GaN barrier and InGaN well and a uniform QW period of GaN/ InGaN. Besides, the RSM analysis demonstrated that the degree of lattice relaxation for sample B was only 7.4 × 10⁻⁵. Both TRPL measurement and µ-PL mapping indicated that the InGaN MQWs exhibited a feeble localization effect, i.e., the InGaN layer in MQWs of sample B has less phase separation and microstructures. Moreover, cross-sectional TEM observations revealed that the epi-layer fabricated on 0.2° off substrate surpasses those fabricated on other substrates in material quality. Hence, the TDD at the bottom of the n-GaN layer is about 9.4 × 10⁷ cm⁻²; however, the TDD at the top of the n-GaN layer reduces to 3.0 × 10⁷ cm⁻², and only 1.0 × 10⁶ cm⁻² in the p-GaN region. Finally, a high-quality $\text{In}_0.2\text{Ga}_{0.8}\text{N}/\text{GaN}$ MQW blue LED was achieved on the 0.2° off substrate, which demonstrated a strong room-temperature EL emission at the wavelength of 465 nm and with a FWHM of only 19 nm. This device also showed a light output of 13.4 mW and an external quantum efficiency of 19.2%. All these characteristics are superior to those of other samples.

Conclusively, the use of sapphire substrates with their c-axis offset by an appropriate offset angle of about 0.2° suggests an effective technique to fabricate high-quality $\text{In}_0.2\text{Ga}_{0.8}\text{N}/\text{GaN}$ MQW blue LEDs.

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