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Effects of Growth Variables on Structural and Optical Properties of InGaN/GaN Triangular-Shaped Quantum Wells

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Abstract−Structural and optical properties of InGaN/GaN triangular-shaped multiple quantum well (QW) structures were investigated under various conditions of growth parameters such as growth temperature, flow rate of Ga and/or In composition, and well and barrier widths. The optical properties affected by the growth parameters were well correlated with an In band gap, which is determined by the potential depth and the In composition in the well region. The emission peak energy was almost independent of the barrier width due to the relaxation of the piezoelectric fields in the triangular-shaped QWs. Photoluminescence spectra of the InGaN/GaN multiple QW structures showed a parabolic curve centered at 2.66 eV. The optical property of the triangular-shaped multiple QWs was substantially improved due to formation of quantum dot-like In composition fluctuations.

Key words: Quantum Well, Quantum Dot, InGaN/GaN, Light-emitting Diode

INTRODUCTION

III-nitrides and their alloys have received much attention due to their tremendous potential for fabricating light-emitting diodes (LEDs) and laser diodes (LDs) that operate in the red to ultraviolet (UV) energy ranges [Strite and Morkoc, 1992; Nakamura, 1998; Pearton et al., 1999; Jain et al., 2000]. InGaN alloy is very important for applications of the III-nitride materials in LEDs and LDs because the alloy constitutes an active region in the form of quantum well (QW) and emits light by recombination of electrons and holes injected into the InGaN active layer. The optical and structural properties of $In_xGa_{1-x}N$/GaN QWs are quite sensitive to the growth conditions of the InGaN layer. However, the quality of InGaN films is dependent on growth variables such as growth temperature, flow rate of Ga and/or In composition, well and barrier widths in the QW regions. Therefore, in order to effectively optimize the growth conditions and tune emission wavelengths, it is necessary to study and understand the effects of growth variables and QW structures on the optical and structural properties of $In_xGa_{1-x}N$/GaN QW structures. Furthermore, understanding the emission mechanism of the $In_xGa_{1-x}N$/GaN QWs is essential for further improving the performance of optical devices.

Several researchers reported the variation of external quantum efficiency (η_{ext}) as a function of emission wavelengths of InGaN-based LEDs [Mukai et al., 1998; Nakamura, 2000; Kaneta et al., 2001]. Although the GaN LEDs emitting at 360 nm exhibited an η_{ext} of approximately 1%, the addition of In (or an increase in In content) to the InGaN active layer resulted in remarkable increase in the η_{ext} value, reaching about 11% at an emission wavelength of 460 nm [Mukai et al., 1998; Kaneta et al., 2001]. However, the η_{ext} decreases if the emission wavelength increases to values greater than the blue-green regions. These phenomena have suggested the recombination of excitons localized at potential minima based on spatial In composition fluctuations, leading to In-rich regions acting as quantum dots (QD) [Narukawa et al., 1997; Arakawa et al., 2000]. However, the crystal quality of the InGaN QWs becomes poor mainly due to the mismatch of lattice constants and the difference in the thermal expansion coefficient between InN and GaN with an increase in the In composition [Ho and Stringfellow, 1996; Romano et al., 1999]. Therefore, in order to improve the η_{ext} of the InGaN-based LEDs and LDs, it is very important to understand and optimize the effects of the growth conditions for the InGaN active layer in terms of structural and optical properties. Recently, we also reported the fabrication of efficient blue LEDs based on the InGaN/GaN triangular-shaped QWs and obtained a substantial improvement of electrical and optical properties of the devices [Choi et al., 2002, 2003, 2004].

In this paper, we report the structural and optical properties of $In_xGa_{1-x}N$/GaN triangular shaped MQWs obtained under various conditions of growth variables. In addition, the emission mechanism of the InGaN QWs is intensively discussed.

EXPERIMENT

Samples used in this study were grown on c-plane sapphire substrates by a low-pressure metal-organic chemical vapor deposition (MOCVD) method. Trimethylgallium (TMGa), trimethylindium (TMIn), ammonia (NH_3), and silane (SiH_4) were used as precursors of Ga, In, N, and Si, respectively. Before growing the nitride films, the substrates loaded into the reactor were thermally cleaned in hydrogen atmosphere at 1,200 °C for 10 min. A GaN nucleation layer of 25 nm thickness was grown on the cleaned substrate at 560 °C, and a 1-μm-thick GaN : Si with 2×10^{18} cm^{-3} of carrier concentration was grown above the buffer layer at 1,130 °C. A 5-period $In_xGa_{1-x}N$/GaN triangular-shaped QW structure was grown on the n-GaN layer under various conditions of growth temperature, flow rate of In, and well and barrier widths. The triangular band struc-

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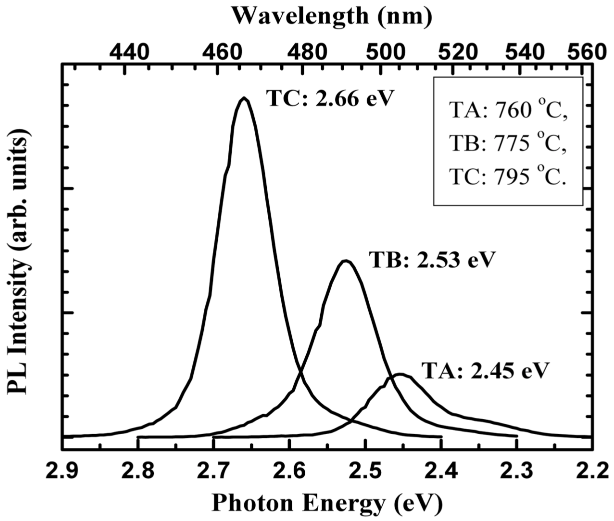

Fig. 1. The PL spectra of InGaN/GaN MQW structures measured at room temperature as a function of growth temperature.

ture in the QWs was obtained by linearly gradating the flow rate of In with time during the growth of the well layer. Details of the growth method are available elsewhere [Choi et al., 2002, 2003, 2004]. A 100 nm-thick undoped GaN capping layer was finally deposited at 1,050 °C on the MQWs. Photoluminescence (PL) was measured with a He-Cd laser operating at 325 nm. Structural properties were analyzed by high resolution X-ray diffraction (HRXRD) and high resolution transmission electron microscopy (HRTEM).

RESULTS AND DISCUSSION

Fig. 1 shows the PL spectra of $In_xGa_{1-x}N$/GaN 5MQW structures grown at 760, 775, and 795 °C, which were named TA, TB, and TC, respectively. The flow rate of TMIn was increased linearly with time up to 32 μmol/min for the first 30 s, maintained constant for 6 s, and then decreased linearly down to zero for the last 30 s. The peak energy linearly decreased to show a red shift with decreasing growth temperature (T_g). The PL intensity also increased with T_g, but full widths at half maximum (FWHM) decreased with T_g. Fig. 2 shows the typical images of cross-sectional high-resolution TEM (top) and bright-field TEM (bottom) images from sample TA. The HRTEM image shows a contrast between the well and barrier, exhibiting five periods of InGaN/GaN QWs. The bright-field TEM image also shows a relatively low threading dislocation (TD) density developed along the c-axis from the underlying layer into the $In_xGa_{1-x}N$/GaN active layer. The TD density estimated from the TEM image is about 1.5×10^8.

Fig. 3 shows the HRXRD patterns of the $In_xGa_{1-x}N$/GaN 5MQW structures of the samples. In principle, the In composition in the films and the period (well and barrier) can be determined by computing the relative shift of the InGaN Bragg peak with respect to the GaN (0002) peak and applying Vegard's law. The period (Λ) is given by

$$\Lambda = \frac{(L_j - L_i)\lambda}{2(\sin\theta_j - \sin\theta_i)} \tag{1}$$

where λ is the wavelength of injected X-ray, L_i and L_j are the orders of satellite peaks, θ_i and θ_j are their diffraction angles, and θ_{0th} is the angle of the 0^{th}-order peak, respectively. Also, higher order diffraction peaks and the FWHMs of satellite peaks indicate high interface quality and good layer periodicity. The strongest peaks are from the GaN epilayer. Based on this and from the HRXRD and HRTEM analysis, the estimated average In compositions of the $In_x Ga_{1-x}N$

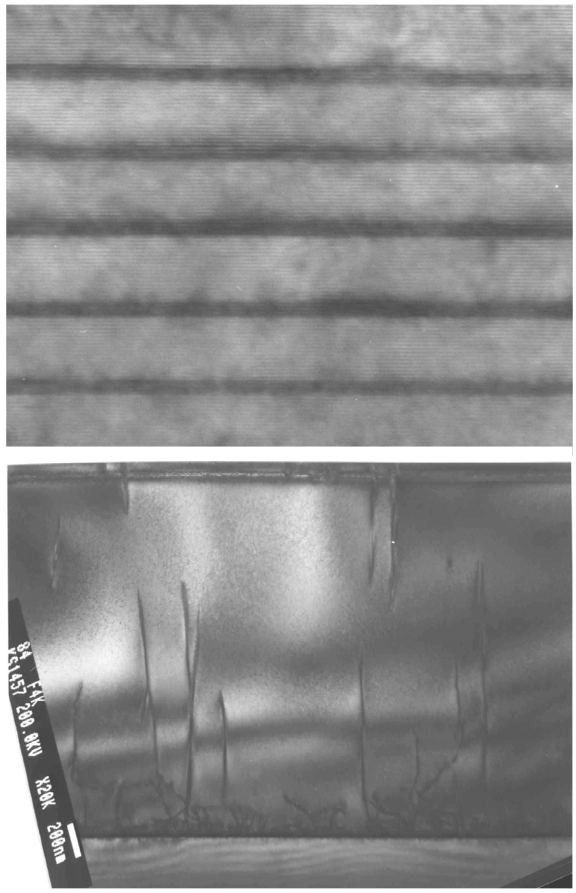

Fig. 2. Cross section of high resolution TEM (top) and cross-sectional bright-field TEM images of InGaN/GaN triangular-shaped multiple quantum well structure (sample TA).

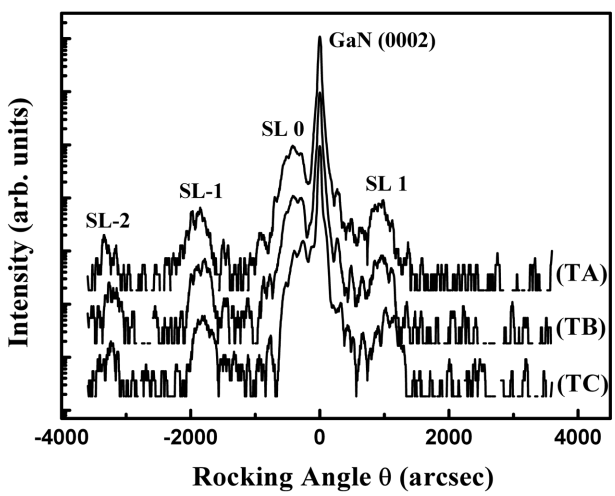

Fig. 3. High resolution XRD patterns of InGaN/GaN MQW structures as a function of growth temperature.

well region are 33, 27, and 20% for samples TA, TB, and TC, respectively. Also, the thicknesses of the well and barrier are 24 and 94 Å for all the samples. The FWHMs of first-order satellite peaks (SL-1) are 266, 254, and 239 arcsec for samples TA, TB, and TC, respectively. That is, the In composition and FWHM of SL-1 increased with decreasing T_g. The amount of In incorporated into the InGaN alloy decreases with increasing T_g due to high volatility of In. Hence, the decrease in the PL intensity and increase of FWHM in the HRXRD at higher In compositions (or at lower temperatures) are presumably due to crystalline imperfection caused by In segregation [Nakamura, 2000; Kaneta et al., 2001; Choi, 2003].

Fig. 4 shows the PL spectra of $In_xGa_{1-x}N$/GaN 5MQW structures measured at room temperature at TMIn flow rates (f_{TMIn}) of 18, 24, and 30 μmol/min for samples IA, IB, and IC, respectively. The growth temperature and the growth time of the well for all the samples were kept constant at 750 °C and 30, 6, 30 seconds, respectively. The peak energy linearly decreased with f_{TMIn} to show a red shift. The PL intensity also decreased, but the FWHMs increased with f_{TMIn}.

Fig. 5 shows the HRXRD patterns for the (0002) reflection from the $In_xGa_{1-x}N$/GaN MQW structures of samples IA, IB, and IC, respectively. From HRXRD and HRTEM, the estimated average In compositions of the $In_xGa_{1-x}N$ well region are 24, 28, and 32% for samples IA, IB, and IC, respectively. Also, the thicknesses of the well and barrier are close to 25 and 94 Å for all the samples. The

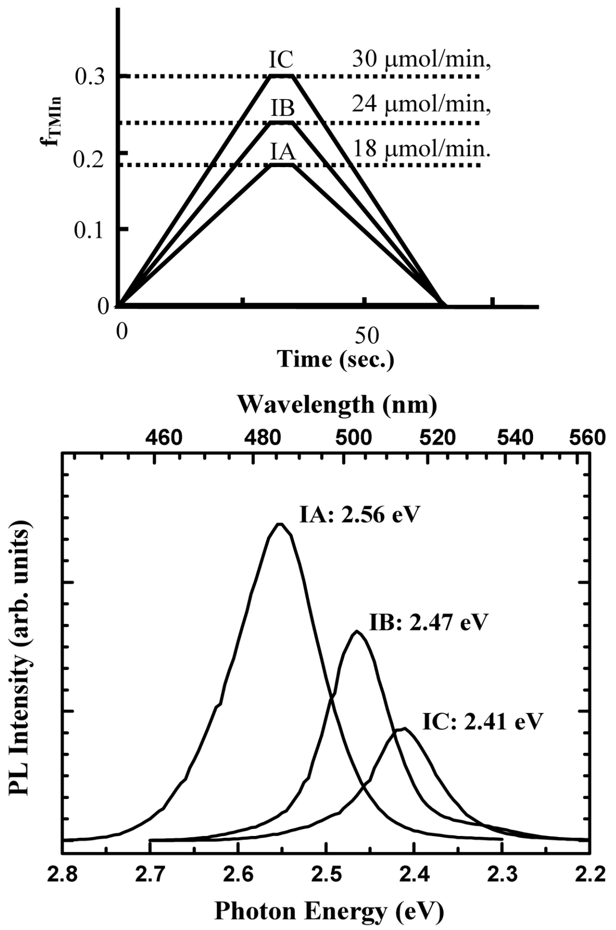

Fig. 4. The PL spectra of InGaN/GaN MQW structures measured at room temperature as a function of flow rate of TMIn.

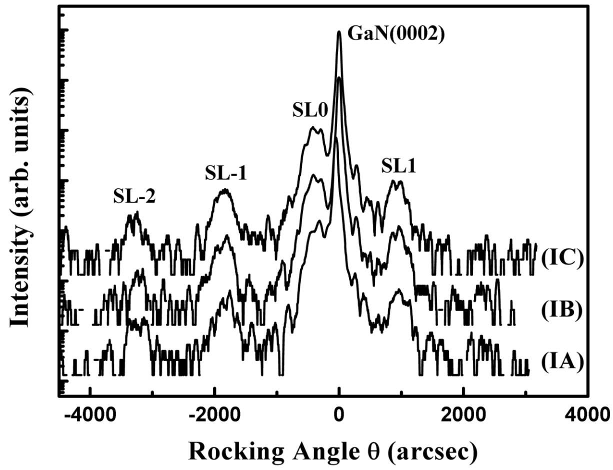

Fig. 5. High resolution XRD patterns of the InGaN/GaN MQW structures as a function of flow rate of TMIn.

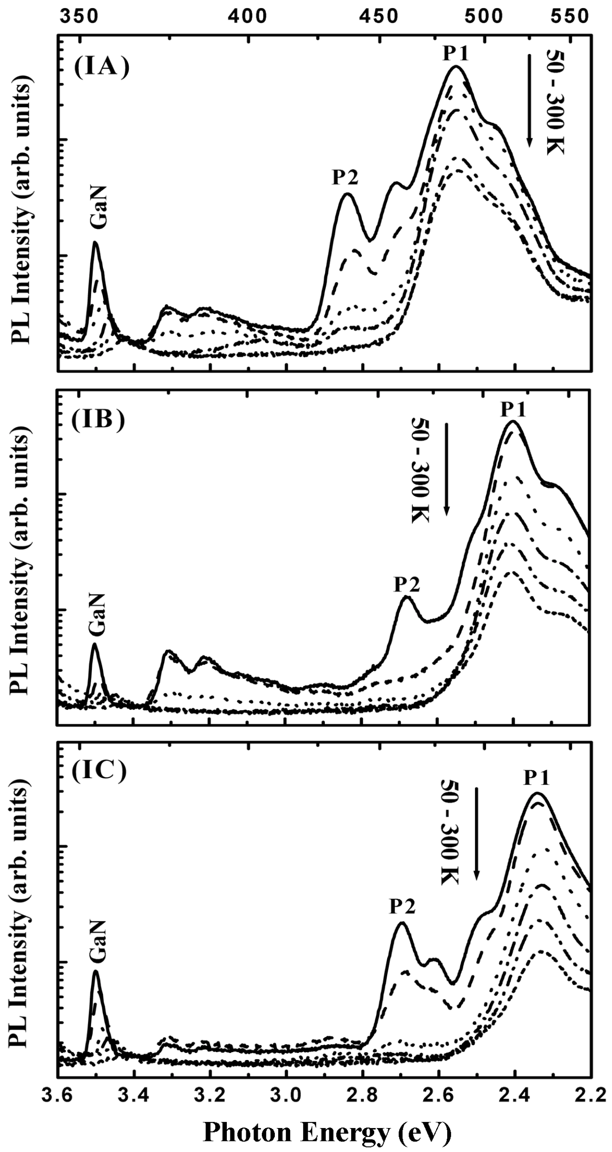

Fig. 6. Temperature-dependent PL spectra of the InGaN/GaN MQW structures grown at various flow rates of TMIn.

FWHMs of SL-1 are 250, 258, and 278 arcsec for samples IA, IB, and IC, respectively. That is, the In composition and the FWHM of SL-1 increased with f_{TMIn}. In general, abruptness at the interface and chemical homogeneity become deteriorated with an increase in the In content due to the difficulty in uniformly incorporating In into the GaN layers [Tran et al., 1998].

To further study the optical property, we measured the temperature-dependent PL of the $In_xGa_{1-x}N$/GaN MQWs in the temperature range of 13-300 K as shown in Fig. 6 (PL spectra at 13 and 30 K are not shown). The PL intensity for all the samples increases with an increase in temperature up to 50 K, and then decreases at temperatures higher than 50 K. This indicates that the radiative recombination process related to excitons localization at potential minima like QDs is dominant at low temperatures, but, as the temperature increases, the non-radiative recombination related to defects caused by crystalline imperfection is prevailing. Two InGaN-related emission peaks (P1 and P2) for all the samples are clearly seen at low temperatures. All the samples also show a less dependence of the peak energy on temperature. As the temperature is increased to 300 K, the degree of degradation of the PL intensity and magnitudes of FWHM variation in the main InGaN-related peak (P1) are increased with f_{TMIn}. These results are presumably due to crystalline imperfection caused by In segregation with increasing f_{TMIn}. Although not shown here, the optical properties of $In_xGa_{1-x}N$/GaN MQWs as a function of growth temperature (T_g) and well width also showed a similar feature as mentioned above. Therefore, for high efficiency green light emission, it is important to effectively incorporate In into the well regions.

Fig. 7 shows the PL spectra of $In_xGa_{1-x}N$/GaN 5MQW structures measured at room temperature versus the growth time (t_w) of the well (i.e., 0, 6 or 12 seconds) for samples WA, WB, and WC, respectively. The growth temperature and the flow rate of TMIn for all the samples were constant at 775 °C and 32 μmol/min, respectively. The peak energy showed a red shift with increasing t_w and the PL intensity decreased with t_w. From HRXRD and HRTEM, the estimated average In compositions and the thicknesses of the $In_xGa_{1-x}N$ well region are 23, 27, 33% and 21, 24, 25 Å for samples WA, WB, and WC, respectively. The thickness of the barrier is 94 Å for all samples. The FWHMs of SL-1 are 243, 254, and 264 arcsec for samples WA, WB, and WC, respectively. Accordingly, the In composition and the FWHM of SL-1 increased with t_w.

Fig. 8 shows the normalized PL spectra of the $In_xGa_{1-x}N$/GaN 5MQW structure measured at room temperature as a function of the growth time of barrier, compared with the triangular (a) and rectangular (b) QW structures, respectively. The triangular-shaped QWs (a) showed stronger intensities and narrower FWHM than those of rectangular ones (b). Furthermore, the peak energy was almost independent of the barrier width in the case of the triangular-shaped QWs, whereas it showed a red shift with an increase in the barrier width in the case of the rectangular-shaped ones. In general, the pie-

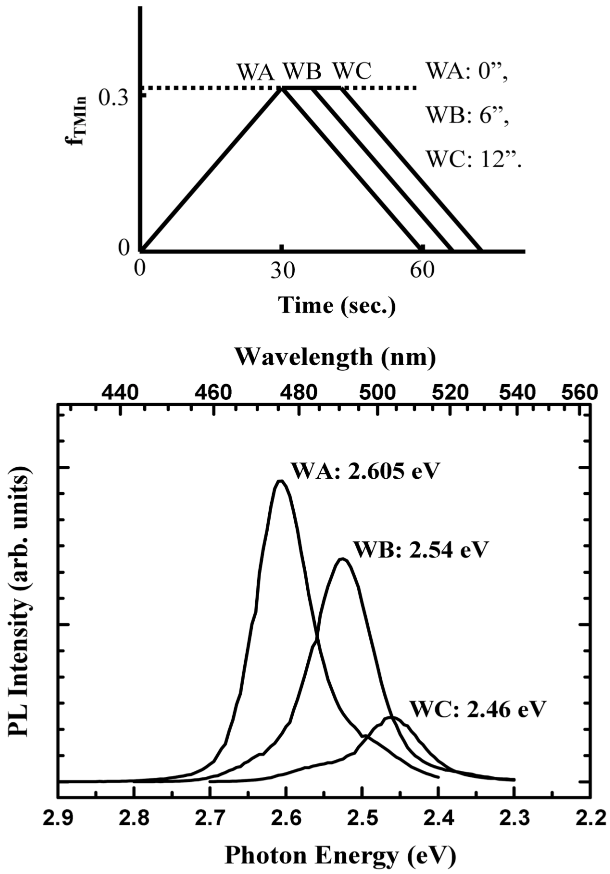

Fig. 7. The PL spectra of InGaN/GaN MQW structures measured at room temperature as a function of well growth time (or width).

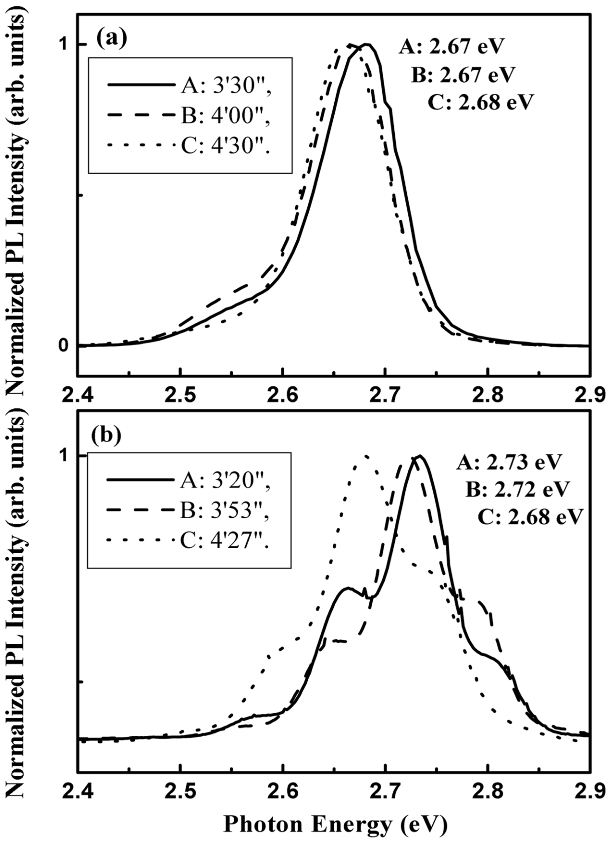

Fig. 8. The PL spectra of InGaN/GaN MQW structures measured at room temperature with barrier growth time (or width); (a) triangular- and (b) rectangular-shaped QWs, respectively.

zoelectric field effects due to the lattice mismatch-induced strained wurtzite InGaN QWs lead to a red shift of luminescence. Also, the electron-hole pair due to the piezoelectric fields are confined to opposite sides of the well, which is expected to yield poor light emission. Therefore, these results are due to the effective relaxation of the piezoelectric fields in the triangular-shaped QWs. Although not illustrated here, the PL spectra of $In_xGa_{1-x}N/GaN$ 5MQW structures grown by varying the emission peak energy (wavelength) from 2.88 to 2.47 eV showed the strongest intensities with an overall parabolic-shape curve centered at 2.66 eV. As we found in our previous works on triangular-shaped QWs [Choi et al., 2002, 2003], an improvement of the optical property in the InGaN/GaN MQWs is attributed to the formation of densely and uniformly distributed In-rich QDs and the relatively low dislocation density.

SUMMARY AND CONCLUSIONS

We investigated the structural and optical properties of InGaN/GaN triangular-shaped MQW structures as a function of growth variables such as growth temperature (T_g), flow rate of Ga and/or In composition, well width, and barrier width. The peak energy showed a red shift with decreasing T_g. The peak energy also showed a red shift as the flow rate of In increased. These results are well described by an In band gap engineering, which manipulates the potential depth by varying the In composition in the well region. However, as the In composition was increased, the PL intensity decreased due to the crystalline imperfection caused by In segregation. The peak energy was almost independent of the barrier width in the triangular-shaped MQWs, but it showed a red shift with an increase in the barrier width in the rectangular-shaped ones. This is probably due to the relaxation of the piezoelectric fields in the triangular-shaped QWs. Also, the PL spectra of $In_xGa_{1-x}N/GaN$ 5MQW structures grown by varying the emission peak energy (wavelength) from 2.88 to 2.47 eV showed strong peak intensities with a parabolic shape centered at 2.66 eV. This is mainly due to the formation of In-rich QDs and the relatively low dislocation density with the triangular-shaped QWs [Choi et al., 2002, 2003]. Therefore, it is necessary for high quantum efficiency to develop a technique capable of effectively incorporating a large amount of In into the well region with sustaining quality films but suppressing phase separation.

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