Structural and Optical Properties of InGaN/GaN Triangular-shape Quantum Wells with Different Threading Dislocation Densities

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Abstract−Structural and optical properties of InGaN/GaN multiple triangular quantum well (QW) structures with different threading dislocation (TD) densities of 1.5×10^{8} (sample A) and 4.5×10^{8} cm^{-2} (sample B) have been studied. High resolution transmission electron microscopy and x-ray diffraction analysis showed more fluctuation of local In composition in the sample B, which was attributed to the stress field created by the dislocations as it provides a driving force for the migration of In atoms towards dislocations. Severe degradation of photoluminescence intensity of the sample B was also observed at >50 K. The optical and structural properties of the InGaN/GaN triangular QW structures are overall substantially affected by the TD density.

Key words: InGaN/GaN, Multiple Quantum Wells, Light-emitting Diodes, Threading Dislocation

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; 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 most commonly used QW structure is a rectangular-type, where In in the well layer of InGaN is generally kept at constant composition. In such a case, spatial indirect recombination is expected due to an internal piezoelectric field formed in the device structure, and thereby poor light emission. Recently, in order to improve device properties, we reported a fabrication of efficient blue LEDs with InGaN/GaN triangular-shape quantum wells and obtained a substantial improvement of electrical and optical properties of the devices [Choi et al., 2002, 2003]. In general, the InGaN active layer includes a large number of threading dislocations (TDs) of 1×10^{8} to 1×10^{12} cm^{-2}, originating from the interface between GaN and sapphire substrate due to a large lattice mismatch [Chichibu et al., 1996; Lester et al., 1995]. Besides, the TDs are considered to be formed as a result of a complex set of interactions including the interface energy, the nucleation density, and the island coalescence [Kapolnek et al., 1995]. Even though numerous studies have investigated the origin of these defects and their effects on the structural [Heying et al., 1996], optical [Garni et al., 1996; Rosner et al., 1997], electronic [Weimann et al., 1998], and morphological properties [Qian et al., 1995] of heteroepitaxial GaN layers, no comprehensive study has been reported in terms of a correlation between structural and optical properties of the InGaN QWs having different TD densities, especially in the triangular QW structures.

In this paper, we report the structural and optical properties of $In_xGa_{1-x}N$/GaN multiple triangular quantum wells (MQWs) with two different densities of TD, which are labeled as samples A and B, respectively. In addition, the emission mechanism of the InGaN QWs is systematically discussed.

EXPERIMENTAL

The samples were grown on c-plane sapphire substrates by a low-pressure metal-organic chemical vapor deposition (MOCVD) system. Trimethylgallium (TMGa), trimethylindium (TMIn), ammonia (NH_3), and silane (SiH_4) were used as the precursors of Ga, In, N, and Si, respectively. Before the nitride films were grown, 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 540 °C, and a buffer layer of 0.5-μm-thick undoped GaN was then grown at 1,180 °C. A 1.5-μm-thick GaN : Si with 1.5×10^{18} cm^{-3} of carrier concentration was grown above the buffer layer at 1,160 °C and named as sample A. For the growth of sample B, the growth processes of the nucleation and buffer layers mentioned above were repeated to make a double buffer layer and the 1.5-μm n-GaN was grown on it. A 5-period $In_xGa_{1-x}N$(2.0 nm)/GaN(8.5 nm) triangular QW structure was grown on the n-GaN layer at 815 °C. The triangular band structure in the QW was obtained by gradating In composition linearly with time in the course of growing the well layer. Details of the growth method are available elsewhere [Choi et al., 2002, 2003]. The emission wavelength was tuned near 465 nm by varying the maximum In content. A 100 nm-thick undoped GaN capping layer was deposited finally at 1,080 °C on the MQW. Structural properties were analyzed by high resolution X-ray diffraction (HRXRD) and high resolution transmission electron microscopy (HRTEM) using a JEOL

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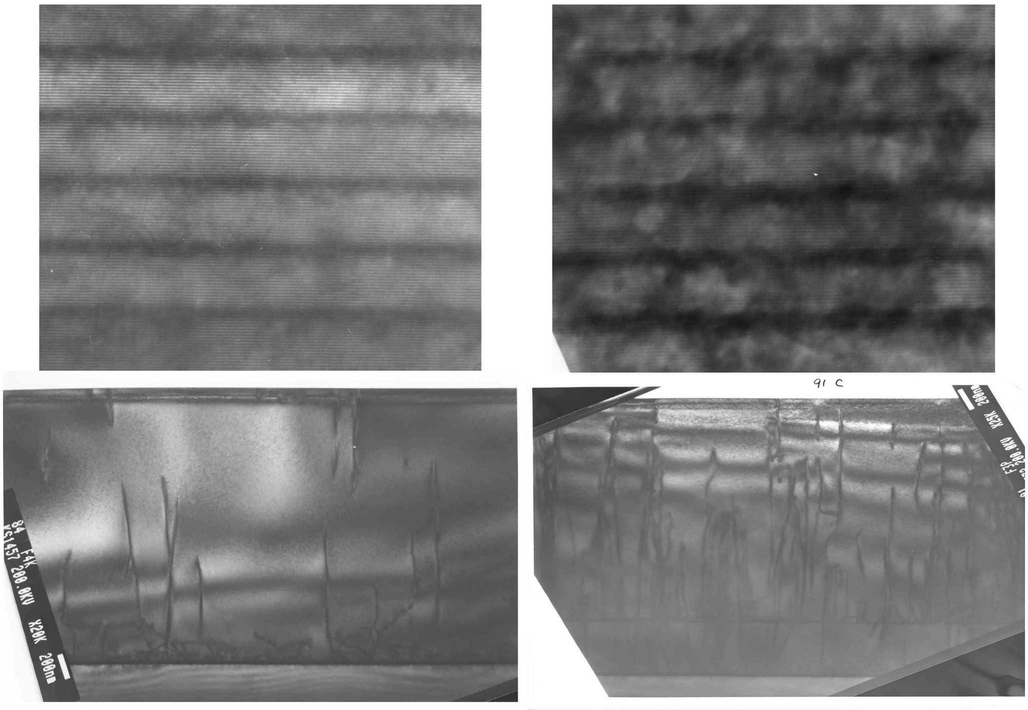
‡This paper is dedicated to Professor Hyun-Ku Rhee on the occasion of his retirement from Seoul National University.



(a) (b)

Fig. 1. Cross sectional high resolution TEM (top) and cross sectional bright-field TEM images (bottom) from the InGaN/GaN triangular MQW structures: (a) sample A and (b) sample B.

2010 operated at 200 kV. Photoluminescence (PL) measurements were carried out as a function of temperature by using a He-Cd laser operating at 325 nm.

RESULTS AND DISCUSSION

Fig. 1 shows the cross-sectional high-resolution TEM (top) and bright-field TEM (bottom) images of the samples A (a) and B (b), respectively. The HRTEM images show a contrast between the wells and the barriers, exhibiting five periods of InGaN/GaN QWs. The variation of contrast, dark area, in the HRTEM image represents the fluctuation of local In composition. Sample A shows quite abrupt interfaces and homogeneous InGaN layers, while sample B shows relatively high In segregations. It is believed that the stress field created by the dislocations provides a driving force for the migration of In atoms toward dislocations. This stress-induced migration may lead to the segregation of In atoms near dislocations and to the formation of In-rich clusters. It is seen from the bright-field TEM images (Fig. 1, bottom) that compared to the sample A, the sample B shows a relatively high TD density developed along the c-axis from the underlying layer into the $In_xGa_{1-x}N$/GaN active layer. The TD densities estimated from the TEM images are 1.5×10^8 and 4.6×10^8 cm^{-2} for sample A and B, respectively. In our previous works on the triangular quantum wells of InGaN/GaN [Choi et al., 2002, 2003], the formation of In-rich quantum dots (QDs) and relatively low dislocation density were observed from a plan-view of TEM image, resulting in a very high blue light-emitting efficiency. The sizes of the QDs in the triangular QWs were in a range of 2-50 nm, which were more densely, uniformly distributed and much smaller than those formed in the conventional rectangular QWs. The QDs were pinned by a dislocation line inferring an interaction between the dislocations and the nucleus of QD formation. We believe that lower TD density and more formation QDs contributed to an enhancement of light-emitting of the InGaN/GaN LEDs, mainly because of the triangular shape of the well layer.

Fig. 2 shows the HRXRD patterns for the (0002) reflection from the InGaN/GaN triangular QW structures with different TD densities (i.e., samples A and B). The strongest peaks are from the GaN epilayer. Both samples clearly show higher order satellite (SL) dif-

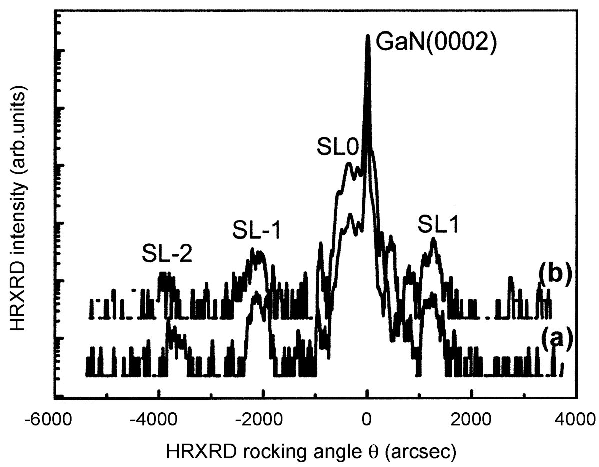


Fig. 2. High resolution XRD patterns of the InGaN/GaN triangular MQW structures: (a) sample A and (b) sample B.

fraction peaks. It is known that the presence of SL peaks indicates good periodicity and abruptness of the QW structures. However, the comparison of the XRD patterns with the HRTEM images (see Fig. 1, top) does not ensure such a deduction. That is, a clear existence of the higher order SL peaks in the XRD patterns, but a poor abruptness with more In fluctuation in the TEM images. In principle, the average In composition and the period can be determined by the relative positions of the 0^{th}-and-higher-order peaks with respect to the GaN peak [Kim et al., 1998; Krost et al., 1995]. Based on this, the In compositions of the $In_xGa_{1-x}N$ wells were 28 and 31% for samples A and B, respectively. The thicknesses of the well and the barrier from the TEM images were 20 and 85 Å, respectively, for both samples. The full widths at half maximum (FWHM) of first-order satellite peaks were 281 and 381 arcsec for samples A and B, respectively, showing a good agreement with the HRTEM images in terms of abruptness. Although not illustrated, the atomic force microscopy (AFM) measurements showed a smaller value of root-mean-square roughness of sample A (0.178 nm) than that of sample B (0.275 nm). These results together with the TEM images lead to a conclusion that dislocations developed from the underlying layer affect the quality of the $In_xGa_{1-x}N$/GaN triangular QW systems.

Fig. 3 shows the PL spectra of $In_xGa_{1-x}N$/GaN 5MQW structures measured at room temperature. Both samples of A and B showed the main peak around 465 nm, but the PL intensity of sample B is almost 50% weaker than that of sample A, mainly because of a higher TD density. Also, the PL linewidth showed 13.6 and 15.3 nm for samples A and B, respectively. Such a better property of the sample A is attributed to the relatively low TD density compared to the sample B. To further study the effect of TD density on the optical property, we investigated the temperature-dependent PL measurement of the InGaN/GaN triangular QWs. Both samples showed the main PL peak at around 465 nm and longitudinal optical phonon replica at long wavelengths. The main peak was attributed to the recombination of excitons localized in QDs formed in the well layer [Choi et al., 2002, 2003]. The peak position and shape were almost independent of the temperature, unlikely in the case of rectangular QWs. PL intensity in both samples increased substantially with the temperature up to 50 K, and then decreased with further increasing the temperature up to 300 K (see Fig. 4). This indicates that carriers are thermally excited to move away from the localized states at temperatures above 50 K. Cho et al. reported that the radiative recombination process related to exciton localization at potential minima is dominant at a low temperature, but at a high temperature, the nonradiative recombination related to defects such as dislocations in the InGaN QW is prevailing. It is also interesting to see that the degree of PL intensity degradation at T>50 K was somewhat greater in sample B than sample A. This is presumably due to the nonradiative recombination process related to more defects in the sample B as shown in the TEM images.

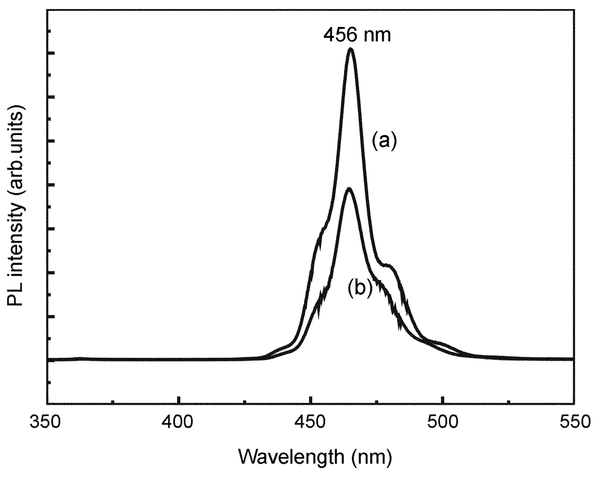


Fig. 3. PL spectra of the InGaN/GaN triangular MQW structures, measured at room temperature: (a) sample A and (b) sample B.

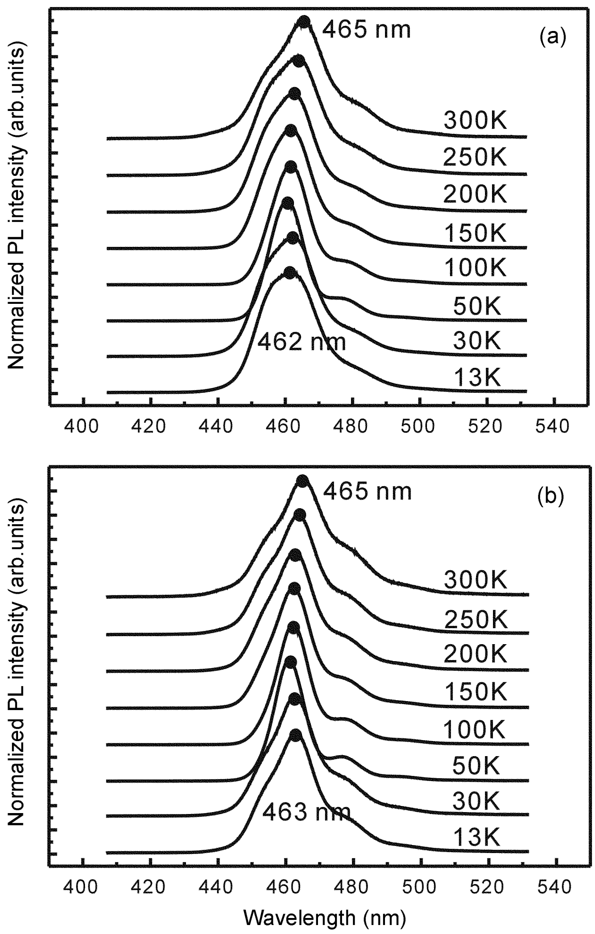


Fig. 4. Normalized integrated PL intensity of the InGaN/GaN triangular MQW structures as a function of temperature: (a) sample A and (b) sample B.

SUMMARY AND CONCLUSIONS

We investigated the structural and optical properties of InGaN/GaN triangular QW structures with different threading dislocation densities. TEM images revealed that the stress field created by the dislocations provides a driving force for the migration of In atoms toward dislocations, resulting in the fluctuation of local In composition in the well layer. Existence of the higher order satellite peaks in the XRD patterns does not guarantee an abruptness of interface in the TEM images. Such a structural analysis led to the conclusion that the dislocations developed from the underlying layer affect the quality of the $In_xGa_{1-x}N$/GaN triangular QW systems. Temperature-dependent PL intensity increased substantially with the temperature up to 50 K, but decreased at higher temperatures (>50 K). More degradation of PL intensity at >50 K was observed with a higher density of threading dislocation. Hence, we deduce that optical and structural properties of the InGaN/GaN triangular quantum well structures are substantially affected by the threading dislocation density.

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REFERENCES

Chichibu, S., Azuhata, T., Sota, T. and Nakamura, S., "Spontaneous Emission of Localized Excitons in InGaN Single and Multiquantum Well Structures," *Appl. Phys. Lett*., **69**, 4188 (1996).

Cho, Y. H., Gainer, G. H., Fischer, A. J., Song, J. J., Keller, S., Mishra, U. K. and Denaars, S. P., "S-shaped Temperature-dependent Emission Shift and Carrier Dynamics in InGaN/GaN Multiple Quantum Wells," *Appl. Phys. Lett*., **73**, 1370 (1998).

Choi, R. J., Shim, H. W., Jeong, S. M., Yoon, H. S., Suh, E.-K., Hong, C.-H., Lee, H. J. and Kim, Y. W., "Triangular Quantum Well of InGaN-GaN for Active Layer of Light-Emitting Device," *Phys. Stat. Sol*., **192**(2), 430 (2002).

Choi, R. J., Shim, H. W., Han, M. S., Suh, E.-K., Lee, H. J. and Hahn, Y. B., "Efficient Blue Light-emitting Diodes with InGaN/GaN Triangular Shaped Multiple Quantum Wells," *Appl. Phys. Lett*., **82**, 2764 (2002).

Choi, R. J., Hahn, Y. B., Shim, H. W., Suh, E. K., Hong, C.-H. and Lee, H. J., "Improvement of Electrical and Optical Properties of InGaN/GaN-Based Light-Emitting Diodes with Triangular Quantum Well Structure," *Korean J. Chem. Eng*., **20**, 1134 (2003).

Garni, B., Ma, J., Perkins, N., Liu, J., Kuech, T. F. and Lagally, M. G., "Scanning Tunneling Microscopy and Tunneling Luminescence of the Surface of GaN Films Grown by Vapor Phase Epitaxy," *Appl. Phys. Lett*., **68**, 1380 (1996).

Heying, B., Wu, X. H., Keller, S., Li, Y., Kapolnek, D., Keller, B. P., Denbaars, S. P. and Speck, J. S., "Role of Threading Dislocation Structure on the x-ray Diffraction Peak Widths in Epitaxial GaN Films," *Appl. Phys. Lett*., **68**, 643 (1996).

Jain, S. C., Willander, M., Narayan, J. and Overstraeten, R. V., "III-nitrides: Growth, Characterization, and Properties," *J. Appl. Phys*., **87**, 965 (2000).

Kapolnek, D., Wu, X. H., Heying, B., Keller, S., Keller, B., Mishra, U. K., Denbaars, S. P. and Spect, J. S., "Structural Evolution in Epitaxial Metalorganic Chemical Vapor Deposition Grown GaN Films on Sapphire," *Appl. Phys. Lett*., **67**, 1541 (1995).

Kim, I.-H., Park, H.-S., Park, Y.-I. and Kim, T., "Formation of V-shaped Pits in InGaN/GaN Multiquantum Wells and Bulk InGaN Films," *Appl. Phys. Lett*., **73**, 1634 (1998).

Krost, A., Bohrer, J., Dadgar, A., Schnabel, R. F., Hansmann, B. S. and Burkhard, H., "High-resolution x-ray Analysis of Compressively Strained 1.55 μm GaInAs/AlGaInAs Multiquantum Well Structures Near the Critical Thickness," *Appl. Phys. Lett*., **67**, 3325 (1995).

Lester, S. D., Ponce, F. A., Craford, M. G. and Steigerwald, D. A., "High Dislocation Densities in High Efficiency GaN-based Light-emitting Diodes," *Appl. Phys. Lett*., **66**, 1249 (1995).

Pearton, S. J., Zolper, J. C., Shul, R. J. and Ren, F., "GaN: Processing, Defects, and Devices," *J. Appl. Phys*., **86**, 1 (1999).

Qian, W., Rohrer, G. S., Skowronski, M., Doverspike, K., Rowland, L. B. and Gaskill, D. K., "Open-core Screw Dislocations in GaN Epilayers Observed by Scanning Force Microscopy and High-resolution Transmission Electron Microscopy," *Appl. Phys. Lett*., **67**, 2284 (1995).

Rosner, S. J., Carr, E. C., Ludowise, M. J., Girolami, G. and Erikson, H. I., "Correlation of Cathodoluminescence Inhomogeneity with Microstructural Defects in Epitaxial GaN Grown by Metalorganic Chemical-vapor Deposition," *Appl. Phys. Lett*., **70**, 420 (1997).

Strite, S. and Morkoc, H., "GaN, AlN, and InN: A Review," *J. Vac. Sci. Technol. B*, **10**, 1237 (1992).

Weimann, N. G., Eastman, L. F., Doppalapudi, D., Ng, H. M. and Moustakas, T. D., "Scattering of Electrons at Threading Dislocations in GaN," *J. Appl. Phys*., **83**, 3656 (1998).