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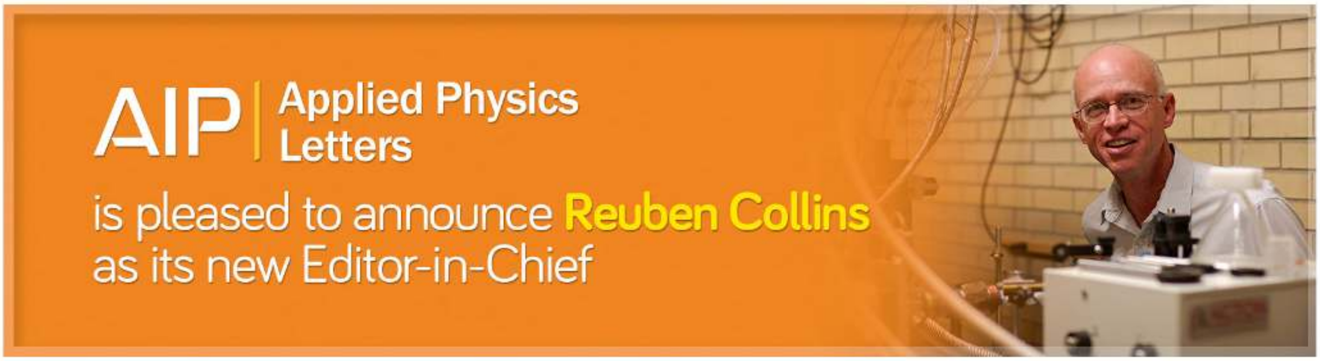

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Optical and microstructural studies of atomically flat ultrathin In-rich InGaN/GaN multiple quantum wells

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Optical and microstructural properties of atomically flat ultrathin In-rich (UTIR) InGaN/GaN multiple quantum well were investigated by means of photoluminescence (PL), time-resolved PL (TRPL), and cathodoluminescence (CL) experiments. The sample exhibits efficient trapping of the photoexcited carriers into quantum wells (QWs) and the effect of internal electric field in the QWs was found negligible by excitation power-dependent PL and TRPL. These phenomena were attributed to the nature of UTIR InGaN QWs, indicating the potential of this system for application in optoelectronic devices. Variation of TRPL lifetime across the PL band and spatially resolved monochromatic CL mapping images strongly suggest that there is micrometer-scale inhomogeneity in effective band gap in UTIR InGaN/GaN QWs, which is originated from two types of localized areas. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2874494]

I. INTRODUCTION

Contrary to conventional Ga-rich InGaN alloys, InN and In-rich InGaN alloys are still at the stage of establishing growth techniques and basic physical properties.[1] It is expected that high defect density would arise from the heterointerface between as-grown InN and GaN since the lattice mismatch between them is larger than 10% and these defects would deteriorate the quality of InN-based heterostructures.[1] Recently, we have overcome these obstacles and have succeeded in growing high quality and atomically flat ultrathin In-rich (UTIR) InGaN/GaN quantum wells (QWs) with In composition of 60%–70% under quite an elevated growth temperature (730 °C) of InN.[2,3] During In-rich InGaN QW growth, only trimethylindium (TMIn) and ammonia (NH_3) were supplied as precursors; however, strain relaxation and solid-state intermixing occurred, and defective In-rich InGaN QW with thickness fluctuation was formed instead of InN QW.[2,3] However, introduction of growth interruption before GaN capping made the formation of defect-annihilated and atomically flat UTIR InGaN layer possible in QWs because of active decomposition and mass transport process in In-rich InGaN layer.[3]

UTIR InGaN QWs seem to be significantly free of internal electric field because of ultrathin thickness and/or high residual carrier concentration in the In-rich InGaN well, leading to internal electric field effect-free optical property. Also, an efficient carrier trapping into UTIR InGaN QWs is expected because excitons in GaN can be effectively localized at the In-rich InGaN well due to the large band offsets between the well and the barrier and smaller electronegativity of In than Ga, resulting in much stronger oscillator strength of excitons, as in the case of InAs/GaAs system.[4] Up to now, little knowledge has been acquired about the optical and microstructural properties of InN and/or In-rich InGaN QWs, unlike conventional Ga-rich InGaN QWs. Here, we report a detailed investigation of optical and microstructural properties of UTIR InGaN/GaN multiple QWs (MQWs). An efficient photoexcited carrier trapping into UTIR InGaN QWs was found and the effect of internal electric field in UTIR InGaN QW layer was negligible by excitation power-dependent photoluminescence (PL) and time-resolved PL (TRPL) experiments. Spatially resolved monochromatic cathodoluminescence (CL) mapping images of the MQW and the variation in PL decay time at various energy levels in the PL band indicated the presence of micrometer-scale effective bandgap inhomogeneity.

II. EXPERIMENTAL

UTIR InGaN/GaN MQW was grown by metal-organic chemical vapor deposition operating at 300 Torr. The sample was grown on a *c*-plane sapphire substrate. The structure consists of a 2-μm-thick GaN buffer layer grown at 1080 °C and ten period InGaN (1 nm)/GaN (20 nm) MQW grown at 730 °C. During the growth of UTIR InGaN QW, only TMIn and NH_3 were supplied as precursors and N_2 carrier gas was

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used. However, strain relaxation as well as solid-state intermixing along growth direction (c-axis) occurred at the early stage of InN deposition on GaN. Medium ion energy scattering (MEIS) measurement showed that the actual In composition in QW layer is 60%–70% and the intermixed regions are present at both top and bottom interfaces.[5] Optical and microstructural properties of the MQW were characterized by PL, TRPL, and CL. PL spectra and decay times of the MQW were obtained at 10 K at various excitation powers by using a second-harmonic light of a picosecond-pulsed Ti:sapphire laser (fixed wavelength: 367 nm, excitation power: 2 mW, and spot size: 400 μm in diameter) and a streak camera with 4 ps resolution. CL experiments were performed using a FEI Quanta 200 scanning electron microscopy equipped with a Jobin Yvon HR460 monochromator and a charge-coupled device camera operating at liquid nitrogen temperature. Typical electron beam voltage and current were 5 kV and 50 pA, respectively.

III. RESULTS AND DISCUSSION

Figure 1(a) shows the 10 K PL spectrum of MQW measured by a He–Cd (325 nm) laser. The near-ultraviolet emission from UTIR InGaN QWs was confirmed by the energy level calculation by Fourier series method using the measured In compositional profile in QWs in our earlier work.[5] Our recent detailed investigation on electronic structures of UTIR InGaN QWs by eight band $k \cdot p$ method showed that the degree of strain relaxation in our QWs does not significantly change the PL transition energy due to relatively strong confinement and proved the accuracy of In composition obtained by MEIS measurement.[6] Figure 1(a) shows that there is almost no luminescence in the GaN band edge position and the integrated PL intensity of GaN band edge is three orders of magnitude smaller than that of QWs. This result strongly indicates an effective bypass of the recombination channels in GaN and the transfer and capture of photogenerated carriers into QWs would be very fast in this system, which is appropriate for light-emitting applications. Also, the internal quantum efficiency of the current structure was evaluated simply by comparing the ratio of the integrated intensity at low temperature and room temperature[7] and it was ~10% at the excitation intensity level of 0.1 W/cm^2. This value seems to be low compared to conventional Ga-rich, thicker InGaN QWs;[7,8] however, internal quantum efficiency is strongly dependent on excitation carrier density[7] and well/barrier strcuture[9] especially in InGaN/GaN system and further study is currently underway on these issues.

We performed PL and TRPL measurements at 10 K to evaluate the effect of internal electric field in the MQW at various excitation powers. The PL spectra showed almost no change in peak position (387 nm) and the normalized line shapes were almost identical when power density was varied from 0.054 to 1.6 W/cm^2, as shown in Fig. 1(b). Furthermore, the TRPL curves showed almost no change as the excitation intensity was varied from 0.054 to 1.6 W/cm^2, as shown in Fig. 1(c). The PL decay times were 1.75 ns, and independent of excitation power and fairly small, compared with previous reports on III-nitride heterostructures.[10,11] In our following study, the electric field effect-free characteristics was also found when we fabricated a light emitting diode structure with UTIR InGaN MQW as an active layer in the current level ranging from 5 to 200 mA.[12] These results suggest that there was scarcely any internal electric field effect in QWs, despite of the existence of high In content of 60%–70% in the well region. It is clear that internal electric field effect weakened with decrease of well width.[13] Although the internal electric field in our QWs would be quite weak due to the ultrathin well width, the significant blueshift of 10–15 nm PL peak is expected with increasing current level to 200 mA from our calculation of eight-band $k \cdot p$ method.[12] However, we could not observe any peak shift with increasing current level. It can possibly be attributed to the high residual carrier density over 10^{18}/cm^3 in In-rich InGaN well, which is enough density to screen the internal electric field.[14,15] Up to now, most of reported residual carrier density in InN is in the order of 10^{18}–10^{21}/cm^3, which is originated from high density of native defects and donor impurities in

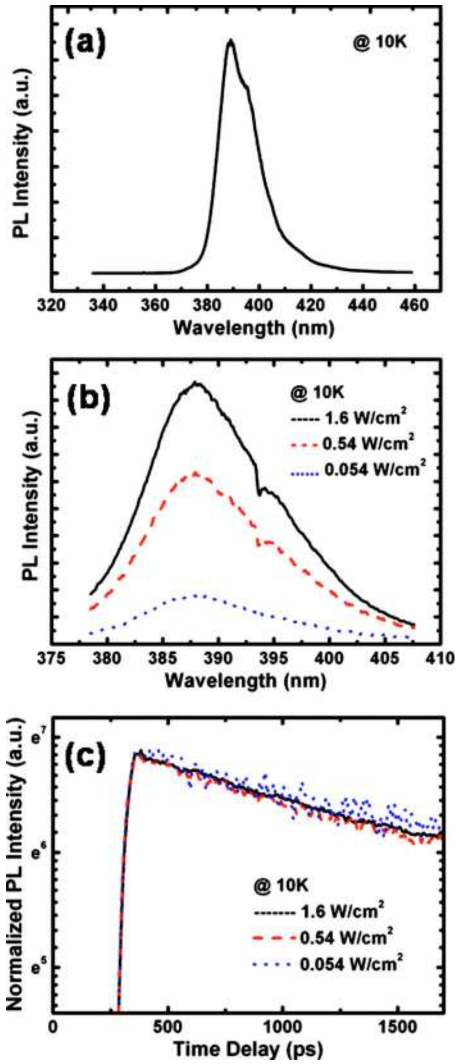

FIG. 1. (Color online) (a) 10 K PL spectrum, (b) excitation power-dependent 10 K PL spectra, and (c) 10 K PL decay curves at 387 nm from ultrathin In-rich InGaN/GaN MQW. The excitation power densities were 0.054, 0.54, and 1.6 W/cm^2 in (b) and (c).

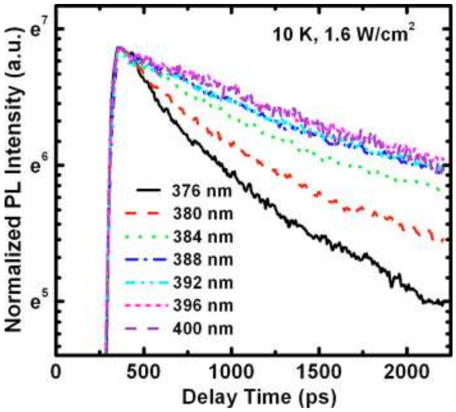

FIG. 2. (Color online) PL decay curves at 10 K at the various energy positions from ultrathin In-rich InGaN/GaN MQW. The excitation power density was 1.6 W/cm^2.

InN (Ref. 1) and we believe that the residual carrier density in In-rich InGaN well would be also in that high range. Consequently, it is expected that our UTIR InGaN QW is perfectly screened by residual carriers, leading to electric field effect-free optical property.

PL decay times were measured across the PL band to study the carrier dynamics in the MQW at the excitation power density of 1.6 W/cm^2, as shown in Fig. 2. Since the measurement temperature was 10 K, the influence of nonradiative recombination processes could be excluded.[16,17] Our experimental data were fitted by a stretched exponential line shape $I(t)=I_1(0)\exp(-t/\tau_1)+I_2(0)\exp[(-t/\tau_2)^\beta]$, which has been used to analyze the emission characteristics of localized system.[16,17] The parameter $I(t)$ means the PL intensity at time t, β is the dimensionality of the localizing centers, and τ_1 and τ_2 are the initial lifetimes of carriers. Both fast lifetime τ_1 and slow lifetime τ_2 increased from 0.88 to 1.75 ns and from 1.60 to 2.49 ns, respectively, as the measurement wavelength was changed from 376 to 400 nm, suggesting carriers trapping at localized states and their transfer from higher energy states to lower energy states.[18]

To confirm the presence of localized states in UTIR InGaN/GaN MQW layer, spatially resolved monochromatic CL mappings were carried out at 5 K for the MQW. From the CL images in Fig. 3, we found that the lateral sizes of each bright area were approximately 0.5–2 μm. The images taken at 376 nm [Fig. 3(a)] and at 406 nm [Fig. 3(f)] were quite complementary to each other. As the emission wavelength increased from 376 to 388 nm, the area of bright region gradually increased. At emission wavelengths around 388 nm [Fig. 3(c)] and 394 nm [Fig. 3(d)], the entire area was bright, which corresponds to the two peak positions in 10 K PL, as shown in Fig. 1(a). These results clearly show that there is micrometer-scale inhomogeneity in effective bandgap in UTIR InGaN/GaN MQWs.

Interestingly, the lateral size of localized centers in UTIR InGaN QWs is much larger than that in conventional Ga-rich, thicker InGaN QWs. To our knowledge, the lateral size of localization centers in most of earlier reports about conventional Ga-rich InGaN QWs has been in the order of a few tens of nanometers and it was explained by the result of indium composition undulation.[19,20] However, in our UTIR InGaN QWs, most of misfit strain in InN was relieved at the very early stage of InN deposition by threading dislocation formation and also by atomic interdiffusion along growth direction (c-axis).[3,5] We believe that the formation of quite large localized centers in UTIR InGaN QWs is rather related with monatomic thickness fluctuation in QWs, not with indium composition undulation because general group-III adatom diffusion length at 730 °C would not be in that high range of micrometer.[21]

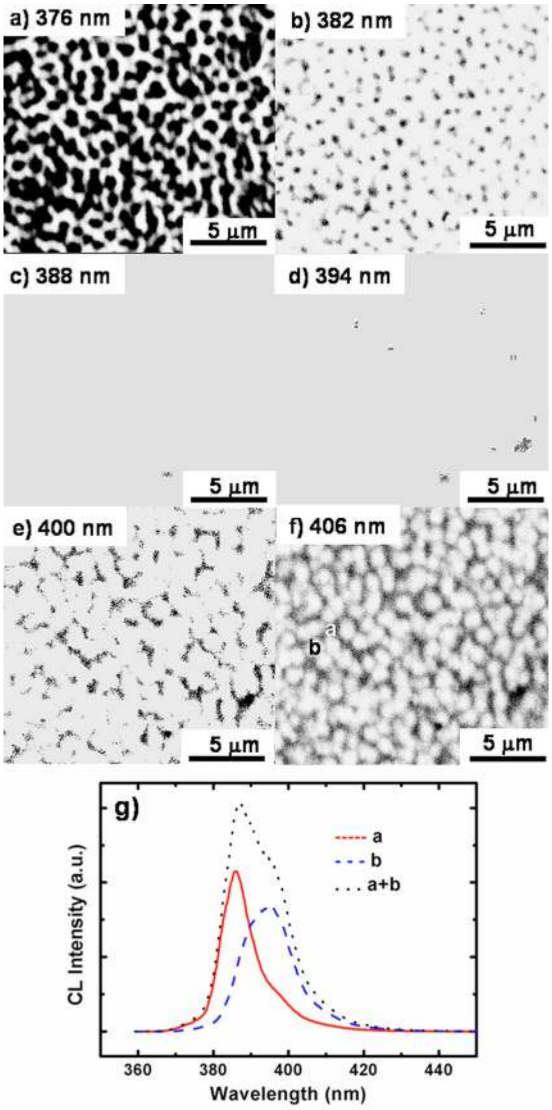

FIG. 3. (Color online) Spatially resolved monochromatic 5 K CL mapping images taken at (a) 376 nm, (b) 382 nm, (c) 388 nm, (d) 394 nm, (e) 400 nm, and (f) 406 nm. The accelerating voltage was 5 kV, indicating luminescence only from MQW region. (g) 5 K CL spectra from different areas of the MQW.

In Fig. 3(f) showing 5 K CL image taken at 406 nm, we denoted dark and bright areas as (a) and (b), respectively. CL spectra obtained from limited spot areas were shown in Fig. 3(g) and the spot CL peak wavelengths in (a) and (b) were 386 and 394 nm, respectively. The spot CL spectra in other areas were also taken from position to position; however, there were little changes in CL peak position. Also, the wide-area integrated CL spectra were almost the same across the whole sample areas. These results can be interpreted as existence of two types of localized areas, resulting in effective bandgap inhomogeneity in QWs and the luminescence from each localized areas added to multiple peaks in CL and PL.

IV. CONCLUSION

In conclusion, the optical and microstructural properties of atomically flat UTIR InGaN/GaN MQW were investigated. We observed an efficient photoexcited carrier trapping into QWs and an electric field effect-free property originated from nature of UTIR InGaN QWs, indicating the potential of this system for light-emitting applications. Micrometer-scale effective bandgap inhomogeneity was present in the QWs and the existence of two types of localized areas was found.

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[1] K. S. A. Butcher and T. L. Tansley, Superlattices Microstruct. **38**, 1 (2005).

[2] S.-Y. Kwon, S.-I. Baik, Y.-W. Kim, H. J. Kim, E. Yoon, J.-W. Yoon, H. Cheong, and Y.-S. Park, Appl. Phys. Lett. **86**, 192105 (2005).

[3] S.-Y. Kwon, H. J. Kim, H. Na, Y.-W. Kim, H.-C. Seo, H. J. Kim, Y. Shin, E. Yoon, and Y.-S. Park, J. Appl. Phys. **99**, 044906 (2006).

[4] M. V. Belousov, N. N. Ledentsov, M. V. Maximov, P. D. Wang, I. N. Yasievich, N. N. Faleev, I. A. Kozin, V. M. Ustinov, P. S. Kop'ev, and C. M. S. Torres, Phys. Rev. B **51**, 14346 (1995).

[5] S.-Y. Kwon, M.-H. Cho, P. Moon, H. J. Kim, H. Na, H.-C. Seo, H. J. Kim, Y. Shin, D. W. Moon, Y. Sun, Y.-H. Cho, and E. Yoon, Phys. Status Solidi A **201**, 2818 (2004).

[6] P. Moon, H. J. Kim, S.-Y. Kwon, E. Yoon, S.-H. Park, and J.-P. Leburton, AIP Conf. Proc. **893**, 391 (2007).

[7] S. Watanabe, N. Yamada, M. Nagashima, Y. Ueki, C. Sasaki, Y. Yamada, T. Taguchi, K. Tadatomo, H. Okagawa, and H. Kudo, Appl. Phys. Lett. **83**, 4906 (2003).

[8] D. M. Graham, P. Dawson, G. R. Chabrol, N. P. Hylton, D. Zhu, M. J. Kappers, C. McAleese, and C. J. Humpreys, J. Appl. Phys. **101**, 033516 (2007).

[9] T. Akasaka, H. Gotoh, H. Nakano, and T. Makimoto, Appl. Phys. Lett. **86**, 191902 (2005).

[10] G. Pozina, J. P. Bergman, B. Monemar, T. Takeuchi, H. Amano, and I. Akasaki, Phys. Status Solidi A **180**, 85 (2000).

[11] P. Hurst, P. Dawson, S. A. Levetas, M. J. Godfrey, I. M. Watson, and G. Duggan, Phys. Status Solidi B **228**, 137 (2001).

[12] H. J. Kim, S.-Y. Kwon, P. Moon, S. Choi, Y. S. Jeon, Y. Moon, E. Yoon, S.-H. Park, T. Chung, J. H. Baek, and Y.-S. Park (unpublished).

[13] T. Kuroda and A. Tackeuchi, J. Appl. Phys. **92**, 3071 (2002).

[14] S.-H. Park and S.-L. Chuang, Appl. Phys. Lett. **72**, 3103 (1998).

[15] J. S. Im, H. Kollmer, O. Gfrorer, J. Off, F. Scholz, and A. Hangleiter, MRS Internet J. Nitride Semicond. Res. **4S1**, 20 (1999).

[16] T. Onuma, A. Chakraborty, B. A. Haskell, S. Keller, S. P. DenBaars, J. S. Speck, S. Nakamura, U. K. Mishra, T. Sota, and S. F. Chichibu, Appl. Phys. Lett. **86**, 151918 (2005).

[17] T. S. Ko, T. C. Lu, T. C. Wang, M. H. Lo, J. R. Chen, R. C. Gao, H. C. Kuo, S. C. Wang, and J. L. Shen, Appl. Phys. Lett. **90**, 181120 (2007).

[18] J. C. Harris, T. Someya, S. Kako, K. Hoshino, and Y. Arakawa, Appl. Phys. Lett. **77**, 1005 (2000).

[19] S. Chichibu, K. Wada, and S. Nakamura, Appl. Phys. Lett. **71**, 2346 (1997).

[20] D. M. Graham, A. Soltani-Vala, P. Dawson, M. J. Godfrey, T. M. Smeeton, J. S. Barnard, M. J. Kappers, C. J. Humphreys, and E. J. Thrush, J. Appl. Phys. **97**, 103508 (2005).

[21] C.-C. Chen, T.-H. Hsueh, Y.-S. Ting, G.-C. Chi, and C.-A. Chang, J. Appl. Phys. **90**, 5180 (2001).