Violet-light spontaneous and stimulated emission from ultrathin In-rich InGaN/GaN multiple quantum wells grown by metalorganic chemical vapor deposition

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We investigated the spontaneous and stimulated emission properties of violet-light-emitting ultrathin In-rich InGaN/GaN multiple quantum wells (MQWs) with indium content of 60%–70%. The Stokes shift was smaller than that of In-poor InGaN MQWs, and the emission peak position at 3.196 eV was kept constant with increasing pumping power, indicating negligible quantum confined Stark effect in ultrathin In-rich InGaN MQWs despite of high indium content. Optically pumped stimulated emission performed at room temperature was observed at 3.21 eV, the high-energy side of spontaneous emission, when the pumping power density exceeds ~31 kW/cm^2. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3002300]

Group III nitrides have been studied widely for their optical and electrical device applications such as laser diodes (LDs), light emitting diodes (LEDs), high frequency electronic devices, and solar cells.[1–4] Especially, InGaN/GaN multiple quantum wells (MQWs) became indispensable commercial solid-state lighting sources in the green/blue/violet spectral regions. In addition, since the InN bandgap was reported as low as ~0.63 eV,[4,5] InN-based III nitrides have recently attracted much attention for extended potential application in a wide range of optoelectronic devices. For the realization of these devices, many groups have studied on the high quality InN and In-rich InGaN nanostructures, including quantum wells[6] (QWs) and quantum dots[7] grown by metalorganic chemical vapor deposition (MOCVD) and molecular beam epitaxy. However, device-quality In-rich InGaN nanostructures have not been obtained yet due to high equilibrium vapor pressure of nitrogen,[8] low dissociation temperature of InN, and large lattice mismatch between GaN and InN (~11%).[6]

In order to resolve the generation of defects and poor interfacial roughness in In-rich InGaN/GaN heterostructures, improvement of optical and structural properties by reducing well thickness and by using growth interruption has been discussed by several groups.[6,9] Recently, Kwon *et al.*[6,10] successfully grew high quality ultrathin In-rich (UTIR) InGaN/GaN MQWs structure with In content of 60%–70% in 1-nm-thick InGaN well on (0001) sapphire substrates at a relatively high growth temperature (730 °C) by MOCVD. Strong near-ultraviolet (UV) emission around 390 nm was observed from UTIR InGaN/GaN MQWs, demonstrating that UTIR InGaN/GaN MQWs structures can be used for the application of near-UV LEDs.[11] However, the study on the stimulated emission (SE) and detailed optical properties of UTIR InGaN MQWs have not been reported yet.

In this letter, we have investigated the spontaneous and SE properties of UTIR InGaN/GaN MQWs in a wide excitation range by means of photoluminescence (PL), PL excitation (PLE), and optically pumped SE experiments.

UTIR InGaN/GaN MQWs were grown on a *c*-plane sapphire substrate by MOCVD at 300 Torr. The structure consists of a 2-μm-thick GaN buffer layer grown at 1080 °C and In-rich $In_xGa_{1-x}N$ (1 nm, $x\sim0.6-0.7$)/GaN (20 nm) with eight period MQWs (sample A) grown at 730 °C. For comparison, we also prepared a 2-μm-thick GaN layer sample without UTIR InGaN MQWs (sample G) and a five-period In-poor $In_yGa_{1-y}N$ (4 nm, $y\sim0.1$)/(In)GaN (10 nm) MQW structure sample (sample B). During the growth of UTIR InGaN well layers only trimethlyindium and ammonia were supplied as precursors, however, InGaN QW with In content of 60%–70% was consequently formed between GaN barriers because solid-state intermixing occurred during InN growth and subsequent growth interruption.[10] More details on growth procedure and compositional analysis of the UTIR MQWs can be found in Refs. 6 and 10.

Figure 1 shows selectively excited PL and PLE spectra of samples A, B, and G measured at 10 K, which were taken by using the quasimonochromatic light from a xenon lamp dispersed by a monochromator. The emission peaks of GaN free exciton for all samples are around 3.485 eV. The sample G shows donor-to-acceptor pair (DAP) transition at ~3.283 eV with its longitudinal optical (LO) phonon replicas. In case of sample B, the PL features corresponding to emission from GaN, (In)GaN barriers (~3.443 eV), and $In_yGa_{1-y}N$ wells are clearly seen along with LO phonon replicas of QW emission. The broad emission at 3.372 eV is probably related to LO phonon replicas or impurity-related emission of (In)GaN layers. The PLE experiments were car-

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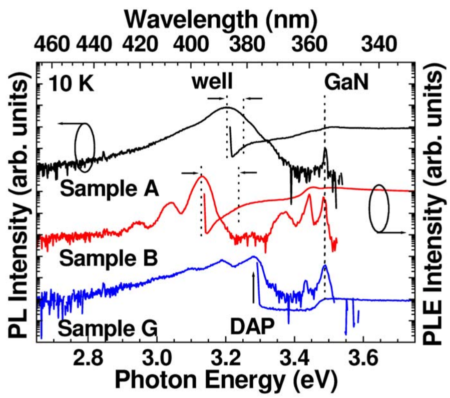

FIG. 1. (Color online) PL and PLE spectra measured at 10 K for samples A, B, and G, respectively. The PLE detection energies of samples A, B, and C were 3.203, 3.128, and 3.283 eV, respectively.

ried out at the detection energies of 3.203 (sample A) and 3.128 (sample B) for InGaN MQWs, and 3.283 eV (sample G) for DAP peak, respectively. A Stokes shift of the InGaN MQWs emission between the PL peak (E_{PL}) and the band-edge (E_B) obtained from the PLE spectra are clearly observed for samples A and B. In order to estimate the Stokes shift, PLE spectra were fitted to the sigmoidal formula of $\alpha = \alpha_0 / \{1+\exp[(E_B\text{-}E)/\Delta E]\}$, where E_B is the average energy gap for the InGaN layer and ΔE is a broadening parameter.[12] We note that the values of E_B were extracted out to be 3.249 (sample A) and 3.233 eV (sample B), respectively. Thereby, the values of Stokes shift ($=E_B\text{-}E_{PL}$) of samples A and B were estimated to be 46 and 105 meV, respectively. In case of sample A, the Stokes shift is extraordinarily decreasing due to ultrathin well layer. We can expect that the reduction in the Stokes shift for sample A indicates an increase in the wave function overlap of electrons and holes, leading to better recombination efficiency of the carriers in the QW.

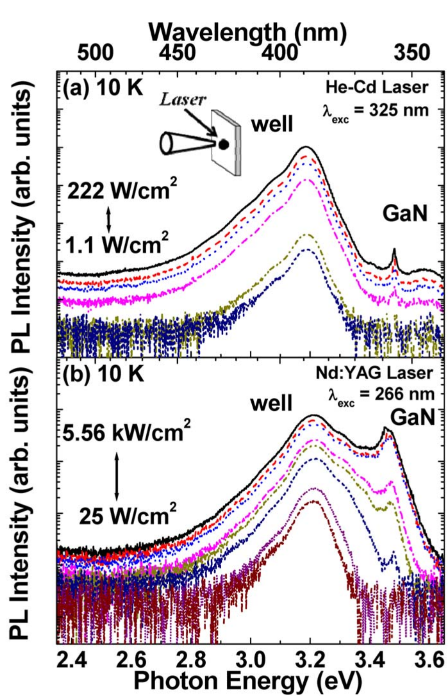

FIG. 2. (Color online) PL spectra of UTIR InGaN/GaN MQWs measured by using (a) a He–Cd laser and (b) a Nd:YAG laser. The excitation power density was varied from 1.1 W/cm^2 and 5.56 kW/cm^2. The inset shows the surface emission geometry.

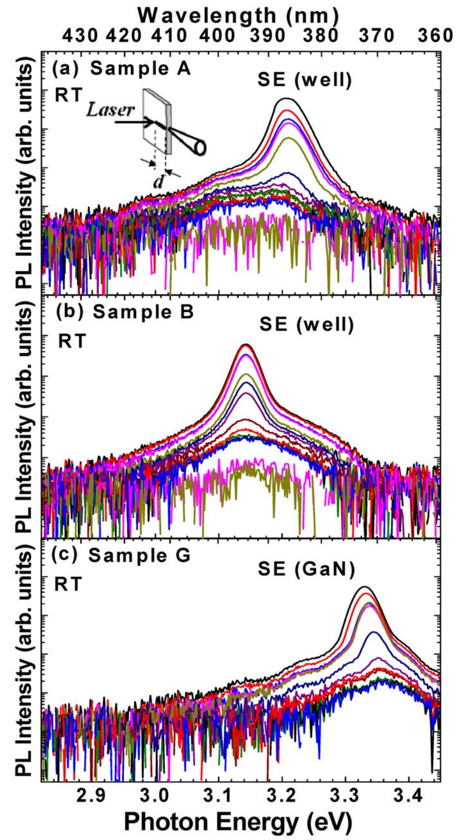

FIG. 3. (Color online) Spontaneous and SE spectra at RT with the edge emission geometry of (a) sample A, (b) sample B, and (c) sample G, respectively. The inset shows the edge emission geometry.

Figures 2(a) and 2(b) show 10 K PL spectra for sample A measured in a wide excitation range by using a 325 nm cw He–Cd laser and a 266 nm pulsed Nd:YAG (YAG denotes yttrium aluminum garnet) laser with a pulse width of 10 ns and a repetition rate of 10 Hz, respectively, which have been performed in surface emission geometry to minimize the effects of reabsorption on the emission spectra, as seen in the inset of Fig. 2(a). Low temperature of 10 K is adopted to rule out the possible redshift caused by heat, and the two laser sources were used to cover power densities over four orders of magnitude from 1.1 W/cm^2 to 5.56 kW/cm^2. This pump power density is supposed to be sufficient for generating enough carriers to result in the screening of the strong internal electric field in the InGaN/GaN grown on (0001) sapphire. Spontaneous emission from the UTIR InGaN/GaN MQWs was observed at 3.196 eV at 10 K and showed almost no change in the peak energy position with increasing pumping power density, as seen in Figs. 2(a) and 2(b). It also indicates that there is negligible quantum confined Stark effect in UTIR InGaN/GaN MQWs despite of containing 60%–70% indium. To explain this behavior, we calculated energy band diagram by eight-band $k \cdot p$ methods and found that the high residual carrier density in high In content InGaN well region over $\sim 10^{18}/\text{cm}^3$ was possibly responsible for the negligible emission wavelength change in UTIR InGaN/GaN MQWs.[13] Detailed energy level calculation using $k \cdot p$ methods and discussion about its effect on the emission wavelength will be reported elsewhere.[11]

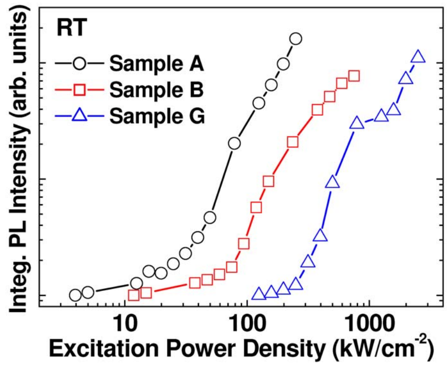

FIG. 4. (Color online) Integrated luminescence intensity as a function of the excitation power density for all samples at RT.

In order to investigate the relevance of In-rich InGaN/GaN MQWs to laser application, optically pumped SE experiments have been performed on all samples at room temperature (RT) in edge emission geometry as seen in the inset of Fig. 3(a). The excitation spot size was approximately $100\times 1000\ \mu m^2$. Figure 3 shows the RT emission spectra of [Fig. 3(a)] sample A, [Fig. 3(b)] sample B, and [Fig. 3(c)] sample G as a function of optical pumping power density ranging from 3.95 kW/cm^2 up to 2.5 MW/cm^2. When pumped by low-excitation power density, the spontaneous emission energies are observed at 3.196 eV (sample A), 3.122 eV (sample B), and 3.359 eV (sample G), respectively. As excitation power density increases, the SE peak appeared at 3.21 eV (sample A) and 3.144 eV (sample B) from InGaN MQWs on high-energy side of spontaneous emission, which is related to the band filling effect of photogenerated carriers and/or compositional fluctuation in InGaN MQWs.[14,15] In case of sample G, however, the SE peak of GaN appeared around 3.349 eV, which is a low-energy side of its spontaneous emission. This is attributed to band gap renormalization due to many-body effects in electron-hole plasma.[16]

Figure 4 shows the plot (log scale) of the integrated luminescence intensities as a function of the excitation power density at the edge emission geometry. A linear increase followed by a superlinear increase in the integrated PL intensity is clearly observed. The threshold power density of sample A is obtained to be $\sim 31\ kW/cm^2$ at RT. This value is two times smaller than that of sample B ($\sim 65\ kW/cm^2$). In addition, we note that the slope efficiency of sample A is also larger than that of sample B, this behavior is expected to exhibit the good device performance. These results are attributed to the electron and hole wave function, which can be more strongly confined in the ultrathin well layer, so that the overlap integral between the electron and hole wave function for sample A is much larger than that of sample B, as mentioned in the PLE results. Although the structures of samples A and B are not exactly the same, we believe that this can be a good comparison between In-rich InGaN MQWs and In-poor InGaN MQWs with different well thicknesses but with similar emission energy. Therefore, we can expect that UTIR InGaN/GaN MQWs structure can be useful for UV spectral region applications such as LEDs and LDs.

In summary, we have studied the spontaneous and SE of UTIR InGaN/GaN MQWs with indium content of 60%–70%. It was found that the large decrease in the Stokes shift in sample A can be attributed to the increase in the overlap of electrons and holes wave function caused by ultrathin well layer. From the RT SE results, this UTIR InGaN/GaN MQW structure has physically and practically significant meaning because this structure can be useful for light sources in UV spectral regions even with high indium content.

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