Stimulated emission study of InGaN/GaN multiple quantum well structures

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We report the study results of an InGaN/GaN multiple quantum well structure with a nominal indium content of 25%. The high-resolution transmission electron microscopy and x-ray diffraction show clear indium aggregation and phase separation. Stimulated emission data always show two major peaks in spectrum. The long- (short-) wavelength peak is assigned to the recombination of localized state carriers (free carriers). At low temperatures or optical pump levels, the localized-state recombination dominates the stimulated emission; however, at high temperatures or pump levels, the free-carrier recombination becomes dominant. The peak position corresponding to localized states changes little in spectrum as temperature or pump level varies. This result is attributed to carrier overflow, strain relaxation, and carrier shielding in increasing temperature or carrier supply. © 2000 American Institute of Physics. [S0003-6951(00)03603-2]

InGaN/GaN quantum well structures have attracted much attention because of their important roles in fabrication of ultraviolet (UV)-blue-green light-emitting diodes and laser diodes. Due to the large lattice constant difference between GaN and InN, indium aggregation and phase separation have been discovered.1–4 Such indium composition fluctuations in InGaN compounds were supposed to be very crucial for efficient light emission. It was believed that such indium-rich nanoscale structures formed the localized states, similar to those of quantum dots. The localized states can trap a significant amount of carriers for radiative recombination, leading to efficient light emission. Therefore, it was widely accepted that the measured photoluminescence (PL) in InGaN samples, and hence the output from a light-emitting diode, came from the recombination of localized excitons.1,5–8 Meanwhile, several stimulated emission (SE) studies have led to the conclusion that their measured SE also originated from band-filled localized states.9–11 Although other research groups reported nonlocalized state laser spectrum,12 it was believed that the gain in laser oscillation was strongly related to the structure of indium composition fluctuation. With such a crucial implication, indium composition fluctuations in InGaN/GaN quantum well structures have received special attention in material and optical studies.

In this letter, we report the results of our optical and material studies on an InGaN/GaN multiple quantum well sample. Two SE spectral components were always observed with their relative intensity varied with temperature and pump fluence. Also, the spectral positions of the SE features, corresponding to the localized states, changed little with the two parameters. The sample studied in this work were grown in a low-pressure metalorganic chemical vapor deposition reactor. The InGaN/GaN multiple quantum well (QW) structure consisted of five periods of silicon doped InGaN well (25% designated indium composition) with 3 nm in thickness. The silicon doping concentration was $10^{18}$ cm$^{-3}$. The barrier was 7 nm GaN. The QW layers were sandwiched with a 1.5 μm GaN buffer layer on a sapphire substrate and a 50 nm capping GaN layer. The growth temperatures were 1050 and 740 °C for GaN and InGaN, respectively. The PL measurements were conducted with a HeCd pumping laser and the SE was pumped (4 mm excitation length) with the fourth-harmonic (266 nm) of a Q-switched Nd:YAG laser (80 Hz).

Figure 1 shows the cross-sectional transmission electron micrograph (TEM) of the sample. The sample was viewed along a (11–20) zone axis. Figure 1 shows the bright field image. The variation of contrast in the micrograph, particularly near quantum wells, represents the fluctuation of indium content. The region of fringe (the upper-center) stands for the occurrence of phase separation or precipitation. Fig-

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\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{figure1.png}
\caption{Cross-sectional transmission electron micrograph of the sample.}
\end{figure}
ure 2 shows the x-ray diffraction result of the same sample, in which InN, GaN, and broadened InGaN features can be clearly seen. From Figs. 1 and 2, one can see quite prominent composition fluctuation in the InGaN alloy.

Figure 3 shows SE spectral variation versus temperature. The alternating solid and dashed curves represent the SE results with temperature varied from 13 K through room temperature (RT). The pump fluence was fixed at 32 mJ/cm². For clarity, we label the three peaks as A, B, and C from the long-wavelength side. The 13 K result corresponds to the highest peak A, the very left peak B, and the highest peak C. As temperature rises, the spectral position of peak A is almost unchanged and its intensity decreases. Also, peak B redshifts and becomes stronger with increasing temperature. Meanwhile, peak C redshifts and becomes weaker with rising temperature. When the temperature is lower than 200 K, there exists a feature between peaks B and C. For comparison, the normalized PL spectra at 13 K (dashed-dotted) and RT (dashed-dotted-dotted) are also plotted. Peak A, which partially overlaps (on the high energy side) with the low-

FIG. 2. X-ray diffraction pattern of the sample.

FIG. 3. SE spectra (solid and dashed curves) of the sample at various temperatures of 13 (LT), 100, 200, and 297 K (RT). The pump fluence was 32 mJ/cm². For comparison, PL spectra at LT (dashed-dotted) and RT (dashed-dotted-dotted) are also plotted.

temperature (LT) and RT PL spectra, is supposed to come from carrier recombination of localized states. Peaks B and C are supposed to originate from free-carrier recombination, corresponding to two different quantum well sub-bands.

The five curves in Fig. 4 stand for the spectral positions of PL (filled triangles), peak A, and peak B, and the peak intensities of peak A and peak B, corresponding to the results in Fig. 3. The redshifts of PL peak and peak B with rising temperature are expected due to band-gap shrinkage. However, peak A position moves little with temperature. Meanwhile, one can see the decreasing and increasing trends of peaks A and B intensities, respectively, with temperature. It is believed that as temperature rises, carriers are thermally excited to move away from the localized states and may occupy free-carrier states (similar to the case of quantum dots). This leads to the decreasing trend of localized state recombination and the increasing trend of free-carrier recombination. The almost unchanged spectral position of localized state recombination (peak A) can also be interpreted for the same reason. Although the global band gap of the material shrinks with temperature, as shown with the PL spectra, mobile carriers with thermal excitation result in almost the same spectral position for the maximum gain. Besides, the weaker strain at a higher temperature in the five quantum well structure, which leads to a blueshift of bandgap, may contribute to cancel the global band-gap shrinkage.

Figure 5 shows the SE spectra with various pump fluences at 13 K. Again, peaks A, B, and C are labeled, the same as those in Fig. 3. The alternating solid and dashed curves stand for different pump fluences with the curve of the highest peak B for the highest pump fluence at 84 mJ/cm² and that of the lowest peak B for the lowest pump fluence at 36 mJ/cm². Both peaks A and B rise with increasing pump level. However, peak A seems to saturate at high pump fluences. At low pump levels, peak A from localized state recombination dominates the emission; however, at high pump levels, peak B from free-carrier recombination becomes dominant. For comparison, the LT PL spectrum is also plotted as the dashed-dotted curve in Fig. 5. Similar to Fig. 4, the four curves in Fig. 6 show the variations of spectral positions...
and intensities of peaks A and B with pump fluence. The redshift trend of peak B with increasing pump level, which is due to band-gap renormalization, was commonly observed. The almost unchanged spectral position of peak A can be attributed to the mutual cancellation between the blueshift, which is due to the filling up of the low band-gap regions and the shielding of piezoelectric field (by strain) with increasing carrier density,\textsuperscript{13,14} and the redshift, which is due to band-gap renormalization. The small change of peak A intensity reflects the filling up of the localized states. The increasing peak B intensity results from the combined effect of increasing carrier supply and the overflow of carriers from localized states as the pump level increases. Although a two-hump SE feature was also observed in an InGaN/GaN multiple quantum well structure, what was reported was the dominance of the short-wavelength hump at low pump levels and the dominance of the long-wavelength hump (due to localized state recombination) at higher pump levels.\textsuperscript{15} This is contrary to what we observed. Note that although the data shown in Figs. 3–6 were obtained with relatively higher pumping levels and a longer excitation length, compared with those in Ref. 15, our similar measurements with the pumping levels near the threshold of SE (with fluence from 6–32 mJ/cm$^2$) and a shorter excitation length (2 mm) showed consistent results. The difference in sample structure, particularly that of indium compositional fluctuation, could be one of the key factors for the different results from Ref. 15.

In summary, we have shown prominent indium aggregation and phase separation in a five-quantum-well InGaN/GaN sample with 25% indium content through the results the TEM and x-ray diffraction. Also, the temperature and pump fluence dependencies of the SE results demonstrated a novel two-peak feature.

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