Emission mechanisms of bulk GaN and InGaN quantum wells prepared by lateral epitaxial overgrowth

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The emission mechanisms of bulk GaN and InGaN quantum wells (QWs) were studied by comparing their optical properties as a function of threading dislocation (TD) density, which was controlled by lateral epitaxial overgrowth. Slightly improved excitonic photoluminescence (PL) intensity was recognized by reducing TD density from $10^{10}\,cm^{-2}$ to less than $10^6\,cm^{-2}$. However, the major PL decay time was independent of the TD density, but was rather sensitive to the interface quality or material purity. These results suggest that TDs simply reduce the net volume of light-emitting area. This effect is less pronounced in InGaN QWs where carriers are effectively localized at certain quantum disk size potential minima to form quantized excitons before being trapped in nonradiative pathways, resulting in a slow decay time. The absence of any change in the optical properties due to reduction of TD density suggested that the effective band gap fluctuation in InGaN QWs is not related to TDs.
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Major developments of III-nitride semiconductors[1,2] have led to the commercial production of blue and green InGaN single quantum well (SQW) light-emitting diodes (LEDs)[1] and to the demonstration of InGaN multiquantum well (MQW) purplish-blue laser diodes (LDs).[1–5] Nakamura *et al.* reported the device lifetime of InGaN MQW LDs more than 10 000 h for cw operation at room temperature (RT) using superlattice cladding layers and nearly threading dislocation (TD) free GaN[4,5] grown by metalorganic chemical vapor deposition (MOCVD) using the lateral epitaxial overgrowth (LEO) technique.[4–10] Although all the high performance LEDs[1] and LDs[1–5] contain InGaN quantum well (QW) active region, the emission mechanisms of GaN[11] and InGaN QWs[1,2,12–16] at RT are not yet fully understood. In particular, a significant decrease of *p-n* junction reverse leakage current in LEO materials was reported[17,18] while Nakamura *et al.*[1,4,5,18] found insignificant change in luminescence intensities of InGaN QW LDs[1,4,5] and LEDs[18] fabricated on LEO GaN, despite that TD density has been reduced by more than four orders of magnitude.[8–10]

In this letter, the spontaneous emission mechanisms in bulk GaN and InGaN QWs are discussed by comparing their optical properties as a function of TD density, which was controlled using the LEO technique during MOCVD growth.

The samples were (i) 1-μm-thick undoped GaN grown on 8-μm-thick coalesced LEO GaN, which was overgrown from 2-μm-thick GaN on sapphire using a SiO_2 pattern with 5-μm-wide openings separated by 15-μm-wide SiO_2 stripes oriented in the $\langle 1\bar{1}00\rangle$ direction. The edge-character TD density in GaN grown vertically from the openings (LEO window) was nearly $10^{10}\,cm^{-2}$ and that in GaN grown laterally over the SiO_2 mask (LEO wing) was less than $10^6\,cm^{-2}$.[10] (ii) 3-nm-thick $In_{0.11}Ga_{0.89}N$ SQWs and (iii) MQWs with five periods of 3-nm-thick undoped $In_{0.15}Ga_{0.85}N$ well and 7-nm-thick Si-doped ($2\times10^{18}\,cm^{-3}$) GaN:Si barriers were grown on the 8-μm-thick LEO GaN. They were capped with 8-nm-thick GaN. In order to extract the optical information of the samples grown on the wing and window regions separately, they were coated with a 150-nm-thick Al layer in which 5-μm-wide stripes were opened to reveal either the wing or the window region. In addition, (iv) 80-μm-thick GaN substrates[5] were prepared.

Time-integrated and time-resolved (TR) photoluminescence (PL) were excited by a frequency-tripled 150 fs pulse from a tunable Ti:sapphire laser. The excitation energy and power at the sample were typically 4.64 eV and 20 mW, respectively.

The PL peak energy of GaN on wing was larger by 15 meV than that of GaN on the window, as shown in Fig. 1. This result suggests that GaN on the wing suffers from larger compressive biaxial strain up to 0.18%[19] after the coalescence. From their peak energies, the PL peaks are assigned to

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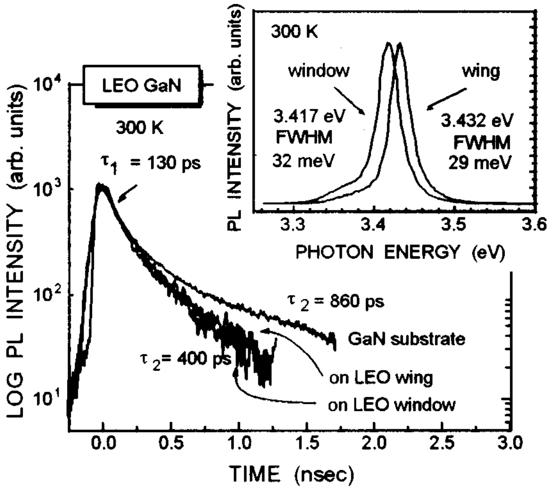

FIG. 1. Time-resolved PL signal from LEO GaN layers grown on the wing and window at RT. For comparison, a signal from 80-μm-thick pure GaN substrate is also shown. Inset shows the PL spectra at RT.

combined A and B free exciton (FE) emission at RT.[11] Generally, RT lifetime is limited by nonradiative processes, and the lifetime of FE emissions in Fig. 1 reflects free carrier lifetime. The TR-PL signal exhibited biexponential-like decay between 4 K and RT. The fast decay (τ_1) was 130 ps at RT and nearly independent of TD density. The time-integrated PL intensity of GaN on the wing was larger than that from GaN on the window, and the dark area distribution in the monochromatic CL images in GaN on window agreed with TD distribution.[20] These results show that TDs act as nonradiative recombination channels. However, the absence of change in τ_1, which dominates the emission intensity, suggests that TDs have little affect on the basic emission mechanisms of GaN. The nonradiative recombination lifetime around TDs is considered to be very short. However, very short diffusion length of carriers in GaN[21] prevents most of carriers from being trapped into TDs. Indeed, the change in the PL intensity of GaN on wing and on window was less than a factor of 10. The lifetime changed remarkably due to change of growth parameters or background carrier density, even if a standard GaN on sapphire with intermediate TD density (5×10^8 cm^{-2}) was analyzed. Therefore the limiting factor of the RT emission intensity is considered to be point defects, impurities, or any electronic nonradiative centers rather than TDs. The pronounced long τ_2 of 860 ps in *pure* GaN substrate may indicate improved purity of the crystal.

The TR-PL decay time measured at the peak energies of InGaN MQWs (2.85 eV) and SQWs (2.97 eV) grown on wing and window does not depend on TD density, as shown in Fig. 2. The InGaN MQWs showed biexponential-like decay, and the lifetime showed very weak temperature dependence.[12,14] Conversely, the lifetime showed a single exponential decay, and the lifetime of InGaN SQWs grown under unoptimized growth conditions decreased rapidly with increasing temperature from 6.4 ns at 10 K to 260 ps at RT accompanied by rapid quenching of the PL intensity, which indicates that PL intensity is limited by nonradiative channels other than TDs.

The long decay time observed in InGaN MQWs can be explained by the combined effects[12] of e-h wave function

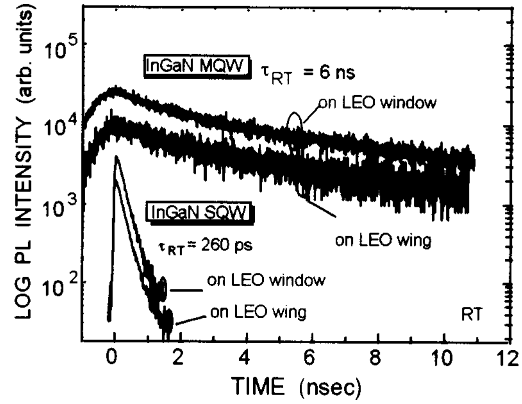

FIG. 2. Time-resolved PL signals from InGaN SQWs and MQWs grown on LEO wing and window regions measured at RT.

separation[12,15,16] due to piezoelectric field[2] and the quantized exciton[12] localization[1,12–14,16] at in-plane potential minima[12] in the QWs. Indeed, the lifetime increases with decreasing emission photon energy, which is characteristic of a localized electronic system, as shown in Fig. 3. The relation between τ and E was fitted using

$$\tau(E)=\frac{\tau_r}{1+\exp[(E-E_{\mathrm{me}})/E_0]}, \qquad (1)$$

where $E_0=60$ meV represents the depth in the tail states, $E_{\mathrm{me}}=2.88$ eV is the energy similar to the mobility edge, and $\tau_r=12$ ns is the radiative lifetime. These values are reasonable for the device-quality InGaN QWs[1]. The low temperature PL lifetime was also independent of the TD density. In addition, the InGaN QWs grown on both the wing and window exhibited the same PL and photoluminesence excitation signals, as shown in Fig. 4. The monochromatic CL images of the InGaN SQW on the window exhibited dimmer areas near the TDs. However, the contrast was much weaker than that observed for GaN,[20] and the overall area was bright. These findings indicate that the lateral diffusion length of carriers in InGaN is shorter than that in GaN and much shorter than the TD spacings.[22] The lateral spacing of the effective band gap minima is considered to determine the carrier diffusion length in InGaN. Note that the diffusion length in strained $In_{0.1}Ga_{0.9}N$ SQWs has been reported to be less than 60 nm.[22] Therefore, such short length scale potential minima can act as a quantum disk (Q disk)[12,23] size po-

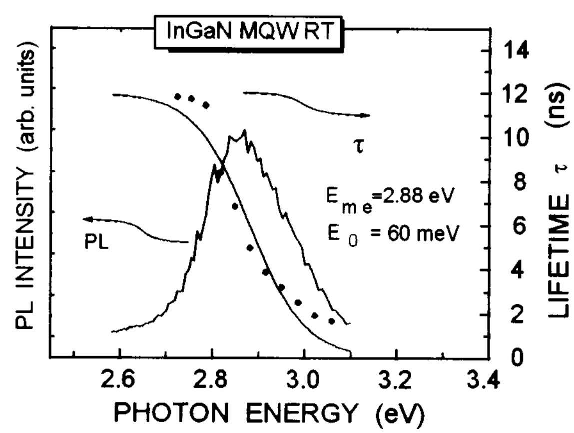

FIG. 3. PL spectra and lifetime of InGaN MQWs grown on LEO wing as a function of emission photon energy measured at RT.

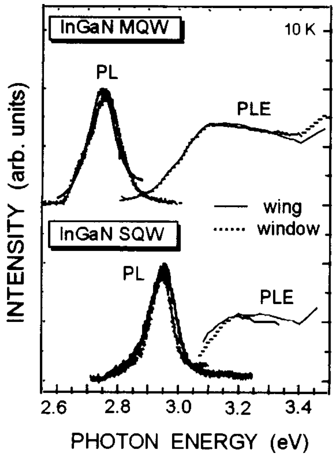

FIG. 4. PL and PLE spectra of InGaN SQWs and MQWs measured at 10 K. No remarkable difference was found in the electronic properties between QWs grown on wing and window.

tential, and it prevents most carriers from being trapped by TDs or any nonradiative pathways between the TDs. This localization can partly explain the improvement of the emission efficiency of InGaN devices[1,3–5,18] compared to that of GaN where carriers are delocalized.

It should be noted that all the optical properties do not depend on TD density. Therefore the in-plane effective band gap inhomogeneity is caused by growth parameters[24] or point defects rather than phase separation initiated by TDs. The Q-disk size/segmented QW potential[12] may, at least partly, come from the alloy potential fluctuation emphasized by the large bowing parameter in InGaN.[25]

In summary, the room-temperature spontaneous emissions were confirmed to be due to recombination of FEs in bulk GaN and of quantized excitons localized at the potential minima in InGaN QWs. TDs act as nonradiative channels for carriers surrounding them within the diffusion length. However, strong localization effectively suppresses the QW excitons from being trapped into TDs or any other nonradiative recombination pathways in the entire area for the case of InGaN, leading to highly efficient emissions from InGaN practical devices. The carrier localization was found to be caused by effective band gap fluctuation initiated by point defects or growth parameters rather than phase separation initiated by TDs. The use of LEO technique actually reduced the TD density from $10^{10}\,\mathrm{cm}^{-2}$ to less than $10^{6}\,\mathrm{cm}^{-3}$, which may improve the LD device lifetime in the form of effective suppression of TD-related degradation mechanisms.

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