

Dependence of carrier localization in InGaN/GaN multiple-quantum wells on well thickness

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Carrier localization in InGaN/GaN multiple-quantum wells (MQWs) with three different well thicknesses was investigated optically using time-integrated and time-resolved microphotoluminescence spectroscopy. An anomalous temperature dependence of the photoluminescence peak energy was observed, as a consequence of local potential fluctuations. The carrier localization was more prominent in the case of MQWs with wide well thickness. The results indicate that the degree of potential fluctuation increases with increasing well thickness. Emission from quantum-dot-like states only became apparent in MQWs with wide well thickness, which supports the assertion that carrier localization in InGaN/GaN MQWs is due to the formation of quantum dots. © 2006 American Institute of Physics. [DOI: [10.1063/1.2423232](https://doi.org/10.1063/1.2423232)]

InGaN/GaN multiple-quantum well (MQW) structures have attracted much attention due to applications in blue light-emitting diode and laser diode devices.^{1,2} Despite high dislocation densities, optical devices exhibit highly efficient luminescence. Although the radiative recombination mechanism is still not fully understood, the quantum-confined Stark effect³ and/or carrier localization effect⁴ are generally accepted as important mechanisms in InGaN/GaN MQW emission. Many proposals have been made as to the origin of localized states in this system. In-rich carrier trapping centers caused by In aggregation and phase separation can lead to a formation of island structures around the InGaN MQW layers, and the In-rich nanoscale clusters surrounded by barriers of high-band gap material act as quantum dots (QDs).⁵ It is well known that the localization of carriers is enhanced with increasing In mole fraction in InGaN layers⁶ in that the amplitude of the potential fluctuations at the band edges is increased by alloy fluctuation between the InN and GaN.

In this work, carrier localization in InGaN/GaN MQWs with the same composition but different well thicknesses will be discussed. We find that the carrier localization becomes stronger with increasing well thickness, which is demonstrated by time-integrated photoluminescence (TI-PL) and time-resolved photoluminescence (TR-PL) measurements. A QD-like state was also observed in micro-PL measurements

for MQWs with wide well thickness. The result suggests that the increase in localization effects is due to the formation of InGaN QDs and a growth model transition from two-dimensional growth for thin layers to three-dimensional growth is promoted with increasing well thickness.

The three MQW samples were grown on *c*-plane sapphire substrates by metal organic chemical vapor deposition. The substrates were initially treated in a H₂ ambient at 1173 °C, followed by the growth of a 25 nm thick low-temperature GaN buffer layer (550 °C) and a 1.5 μm thick layer of nominally undoped GaN grown at high temperature under low pressure. The temperature was then lowered to 742 °C to grow a ten period In_{0.11}Ga_{0.89}N/GaN MQW under 200 torr pressure. All samples were grown in rapid succession under identical conditions except for the well thickness. In all cases the barrier thickness was 7.5 nm, and the well thicknesses were 1.4 nm, 2.4 nm, and 3.9 nm for samples A, B, and C, respectively. A Bede D1 system, a commercial high-resolution x-ray diffraction (XRD) system, was used to perform the structural characterization on all the samples. XRD measurements in ω -2 θ scan and high-resolution mode were performed and a simulation based on XRD dynamic theory was made to fit the experimental curve. Taking sample B as a representative case, the well thickness was found to be 2.4±0.2 nm and the indium mole fraction was 11%±2%.⁷

In order to investigate whether QDs were formed, an optically opaque metal mask with square apertures with side

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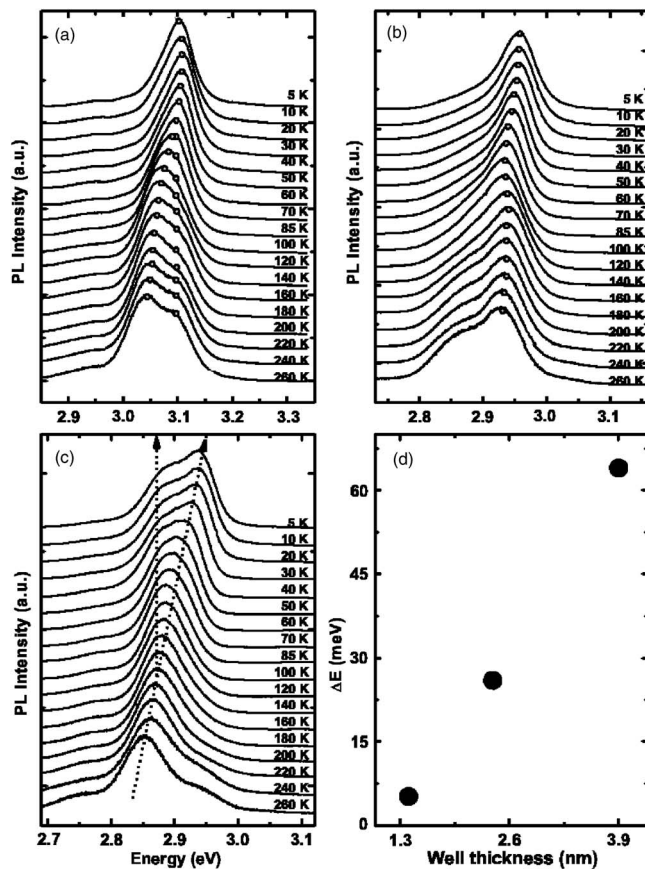


FIG. 1. Temperature-dependent PL spectra of sample A (a), sample B (b), and sample C at temperatures ranging from 5 to 260 K. The power density was 126 W cm^{-2} . The gray circles and dotted lines indicate the evolution of PL peak energies. (d) Energy difference between the $e1-h1$ transition and localized state vs well thickness.

length between 100 nm and $2 \mu\text{m}$ was employed, a typical process for single-QD spectroscopy.

For TI-PL measurements, a HeCd laser at 325 nm was used for excitation, while for TR-PL measurements, frequency-tripled femtosecond Ti:sapphire laser pulses operating at 275 nm were used. A $36\times$ reflective objective was held above the cryostat to both focus the incident laser beam to a spot size of $\sim 2 \mu\text{m}$ and collect the resulting luminescence. For TI measurements the PL was detected by Peltier-cooled charge coupled device, and for TR measurements the PL was directed to a time-correlated single photon counting system using a Si avalanche photodiode detector with a time resolution of 50 ps.

Temperature-dependent TI-PL spectra of the three InGaN/GaN MQW structures in the range from 5 to 260 K are presented in Figs. 1(a)–1(c). The laser excitation power was $15.9 \mu\text{W}$. At 5 K, the PL spectra peak at 3.109 eV, 2.957 eV, and 2.941 eV for samples A, B, and C, respectively, corresponding to transitions between the first electron and first heavy-hole subbands ($e1-h1$).⁸ While the $e1-h1$ transition is dominant for samples A and B at 5 K, for sample C a low-energy shoulder peaked at 2.881 eV was seen together with the $e1-h1$ transition at 2.941 eV. All the samples show an anomaly in the emission shift with temperature. An “S-shaped” shift as a function of temperature is usually observed in InGaN/GaN MQWs and ternary compounds, arising from local potential fluctuations caused by inhomogeneous alloy distribution.^{9,10} The PL peak energy for

sample A blueshifts in the temperature range of 5–20 K, redshifts for 20–70 K, and then separates into two lines above 70 K. It is worth noting that the PL peak at 3.104 eV remains up to 260 K without emission shift and the other peak keeps redshifting, as shown in Fig. 1(a). The PL peak at 2.957 eV for sample B, on the other hand, redshifts to 2.933 eV from 5 to 85 K, blueshifts slightly by $\sim 5 \text{ meV}$ between 85 and 140 K, and then the PL peak shows no emission shift above 140 K, as seen in Fig. 1(b). The low-energy tail seen in sample B probably arises from defect states. It has no relevance, however, to the analysis below. For sample C, the 2.941 eV line redshifts progressively as temperature increases. In contrast to the 2.941 eV line, the 2.881 eV line shows no variation in emission energy for temperatures ranging from 5 to 180 K. The 2.941 eV line merges with the 2.881 eV line at 180 K, as shown in Fig. 1(c). Detailed recombination dynamics for the samples will be reported elsewhere.

Since, in InGaN/GaN MQW structures, photogenerated carriers are transferred from their first quantized states to lower localized centers,^{10–12} we propose that the peaks which do not show a shift with increasing temperature (i.e., peaks at 3.104, 2.933, and 2.881 eV) originate from localized states in each sample. The energy differences between the $e1-h1$ transitions at 5 K and the recombination of localized excitons were 5 meV, 24 meV, and 60 meV for samples A, B, and C, respectively, indicating that the degree of localization (effective potential fluctuations) in InGaN/GaN MQWs increases with increasing well thickness. In order to test this hypothesis, the full width at half maximum (FWHM) of PL spectra recorded at 5 K for all three samples were fitted using single Gaussian functions. The FWHM of the PL spectra increased from $56 \pm 0.2 \text{ meV}$ for sample A to $65 \pm 0.4 \text{ meV}$ for sample B. The increase in the FWHM arises principally from a broadening on the low-energy side of PL spectra. As shown in Fig. 1(c), the recombination at localized states is more dominant in sample C than in sample A or B.

In general, thermal quenching of PL spectra stands for the depth of the confining potential that needs to be overcome by the localized carrier in order to escape and diffuse to a trap. Thus, the thermal activation energy in InGaN/GaN MQWs corresponds to the magnitude of effective potential fluctuations. The activation energies for the samples were deduced from Arrhenius plot of PL intensity as a function of temperature. Activation energies of 27.1 ± 2.3 and $31.0 \pm 2.0 \text{ meV}$ from samples A and B were obtained. Although it is difficult to determine the activation energy of sample C, due to the superposition of the two peaks, the activation energy is greater than 35 meV.

Figures 2(a)–2(c) depict TR-PL spectra monitored at the PL peak energies for the MQW samples under an excitation power of $50 \mu\text{W}$ at 5 K. Such a two-component PL decay in InGaN/GaN MQWs was previously observed by Feng et al.,¹³ the first decay component being attributed to exciton relaxation of free carrier and localized states and the second decay arising from the relaxation of localized excitons. Here, the fast and slow decay components lie in the range of 1.5–2 and 4–5 ns, respectively. According to a carrier localization model,⁴ deeper localized states have longer lifetimes because of stronger localization effects.^{4,14} PL decay times of localized excitons were obtained by fitting the slow decay component, resulting in decay times of 3.260 ns, 9.136 ns, and 29.78 ns for samples A, B, and C, respectively. The PL decay

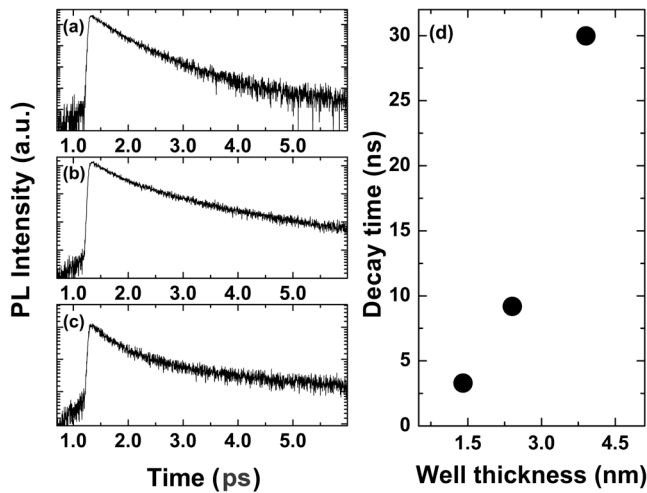


FIG. 2. TR-PL decay curves of samples A (a), B (b), and C (c) recorded at 5 K. (d) Decay time of localized excitons (slow decay component on the biexponential curves) vs well thickness, showing an increase in the decay time with well thickness.

time becomes longer in MQWs with wider well thickness as expected, due to the quantum confined Stark effect.

It is well known that the degree of local potential fluctuations increases with alloy content in ternary compounds.^{14,15} However, these results point out that an increase of potential fluctuations can occur with well thickness as well. Previously, several reports suggested that the localized states may arise from In compositional fluctuation or QD formation, phase separation, monolayer thickness fluctuation, and surface segregation.^{16–18} Figure 3 shows time-integrated macro-PL spectra for the unmasked MQW samples together with a micro-PL spectrum for the Al masked sample C recorded at 5 K (inset) at an excitation power of $15.9 \mu\text{W}$. The micro-PL spectrum shows QD-like emission on the high-energy side of macro-PL spectrum with a FWHM of 1.65 meV. While sample C exhibited clear QD emission (seen in several masked areas), we were not able to

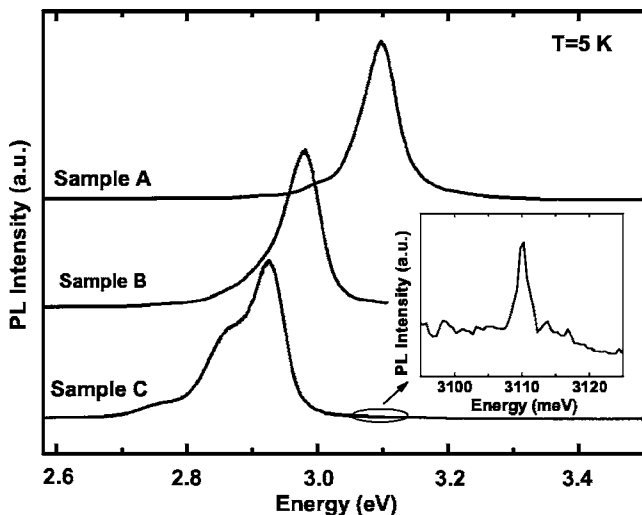


FIG. 3. TI-PL spectra for samples A, B, and C at 5 K. The power density was 126 W cm^{-2} . The inset displays a single QD emission from sample C with a 400 nm square aperture, showing a QD-like discrete energy spectrum. Note that no QD-like PL emission was observed in samples A and B.

detect any QD emission from samples A and B and saw no significant spatial variation in the PL intensity. We suggested previously the presence of QDs in MQWs with wide well thickness, resulting from a growth model transition with increasing well thickness.⁸ Although the QD-like emission was observed only on the high-energy side of PL spectrum, a wide range of potential fluctuations may occur in sample C. However, these deep localized states could not be detected as they would be embedded in the strong QW-related emission. It is therefore clear that potential fluctuations are promoted with increasing well thickness and the origin of enhancement in carrier localization is probably the formation of InGaN QD-like states (i.e., In-rich clusters).

We have investigated carrier localization effects in InGaN/GaN MQWs with different well thicknesses using TI- and TR-PL spectroscopy. In the case of MQWs with a 1.4 nm well thickness, the $e1-h1$ transition is the dominant process. With increasing well thickness exciton localization effects become dominant, implying a greater amplitude of potential fluctuation in wider wells. This strong carrier localization is a consequence of the growth mechanism by which QDs are formed.

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