Time-Resolved Photoluminescence Measurement of Frenkel-type Excitonic Lifetimes in InGaN/GaN Multi-quantum Well Structures

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Time-resolved photoluminescence from InGaN/GaN multi-quantum well structures was investigated for two different shapes of square- and trapezoidal wells grown by metal-organic chemical vapor deposition. To compare to the conventional square well structure with a radiative recombination lifetime of 0.170 nsec, the large value of lifetime of 0.540 nsec from trapezoidal well were found at room temperature. This value is similar to the value for GaN host material indicating no confinement effect of quantum well. Furthermore, the high resolution transmission electron microscopy image provides the In clustering effect in the trapezoidal well structure.

Keywords : Oscillator strength, InGaN/GaN, Quantum well, Photoluminescence

1. INTRODUCTION

For opto-electronic applications, the quantum engineering of InGaN alloyed structures was extensively studied as the active region for short-wavelength visible light emitting devices. The InGaN active layer provides the distinct radiative recombination process due to the highly ionic covalence in the bonding character of GaN-based materials. However, this ionic character provides the strong strain-induced piezoelectric field, which causes a severe distortion of the InGaN/GaN interfacial band structure[1-4].

The dynamic behavior of carriers in the InGaN/GaN superlattice structure can be determined by the implementation of the time-resolved photoluminescence (TRPL) that is a powerful experimental tool to study spectral information[5,6]. The low-temperature decay times for the donor bound exciton(DX) and free A exciton at 5 K were 0.136 and 0.066 nsec, respectively, in the unintentionally doped GaN grown by metal-organic chemical vapor deposition (MOCVD)[7]. However, the DX and the free A exciton have the radiative lifetimes of 0.530 and 0.295 nsec, respectively at 4 K, in the unintentionally doped GaN grown by hydride vapor phase epitaxy (HVPE)[8]. This large discrepancy of lifetimes is closely related to the crystalline quality and the photoluminescence center is very sensitive to the micro-structure. This useful information of TRPL dependency on the crystalline quality can be extended into the optimization of quantum well structures as a characterization probe.

In this work, we introduce trapezoidal and square InGaN/GaN multiple quantum well structures grown by MOCVD in order to compare the reduction of the piezoelectric field effect and present the results of time-resolved studies of photoluminescence. The radiative recombination lifetime of 0.170 nsec from square well and 0.540 nsec from trapezoidal well were found at room temperature corresponding to the oscillator strengths of 3 and 1.1, respectively. The large value of the lifetime for trapezoidal well implies the structural relaxation.

2. EXPERIMENTAL

InGaN/GaN multiple quantum wells(MQW) were grown in a horizontal MOCVD reactor operating at pressure of 400 Torr. The nucleation layer on sapphire substrate was deposited at the temperature of 560 °C and then the Si-doped GaN film was grown at the temperature of 1130 °C with the thickness of 2 μm. Subsequently, the six periods of InGaN/GaN quantum well structures were fabricated at 795 °C for the trapezoidal and square wells with and without the In compositionally graded region, respectively. The In mole fractions of 0.23 and 0.25 for the trapezoidal and the square wells were, respectively, estimated by the double crystal X-ray diffraction(DCXD). The average widths of wells are 2.5 nm-thick and the barriers are 9-10 nm.

Time-resolved photoluminescence measurements are performed using a pulsed pico-second mode-locked Nd:YAG laser (Coherent Inc.) and a micro-channel plate photo-multiplier tube (MCP-PMT) (Hamamatsu R3809-U-51). The dual-jet dye laser pulses are frequency-doubled resulting in an operation wavelength region at 570-610 nm. The repetition rate is changed to 13.66 MHz using a Pockel cell. The average power is 40 mW and the typical diameter of the laser spot on the sample is 100μm. The emitted light is dispersed by a monochrometer (HR320 with 1200 (grooves/mm) grating (Join-Ybon) and collected onto the photocathode of the MCP-PMT. The PL data are recorded by a PC interfaced time-correlated single photon counting (TCSPC). The measurement system had a resolution of 20 ps.

3. RESULTS AND DISCUSION

Figure 1 shows the room temperature PL spectra from the samples of square and trapezoidal well structures. The intense PL peaks of 479 and 495 nm were observed from the sample of square well structure. However, the relatively weak PL peak at a shorter wavelength of 457 nm was observed from the sample of trapezoidal quantum well structure. The energy level in MQW is quite dependent on the size of the well width and the shape of the well. The In compositional grading at the well/barrier interface provides the change of the shape of well and also the energy level. The direct comparison of two types of well shape is a very delicate problem.

Strictly speaking, the PL results may include the piezoelectric field effect on band structure at the vicinity of the hetero-interface and provide the red-shift in square well structure rather than trapezoidal well structure. The PL intensity from the square multi-quantum well structure is much stronger than from the trapezoidal MQW structure indicating the structural morphology of later structure is not better than the former structure by involving with structural non-uniformity such as In clustering.

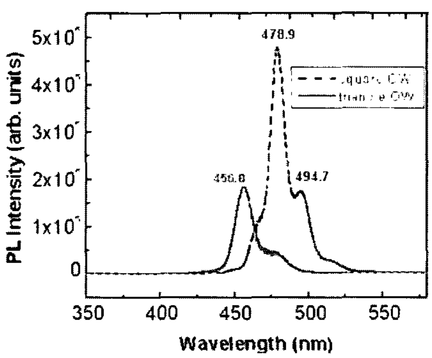

Fig. 1. The room-temperature PL spectra from square and trapezoidal well structures.

Table 1. Recombination lifetime constants in two exponential decays.

QW type	PL (nm)	A_1	A_2	τ_1(ns)	τ_2(ns)	f_1
Square	479	0.4	0.35	0.17	0.65	4.06
	495	0.36	0.27	0.33	0.79	2.09
	515	0.44	0.16	0.45	1.01	1.53
Trapezoid	457	0.35	0.22	0.54	1.80	1.27
	479	0.28	0.25	0.58	1.70	1.19

Figure 2 shows the TRPL spectra measured on the sample of square well structure at room temperature with a fixed pump power of 40 mW. The TRPL spectra were fitted into the combination form of two exponential decays and recombination lifetimes were obtained as shown in Table 1. The TRPL spectra were measured at the three optical interference positions of PL peaks at 479, 495 and 515 nm. The two exponential fits have been used by the following form[9]:

$$I(t) = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2) , \qquad (1)$$

$$\tau = \frac{21.548}{E_g{}^2 \, n \, f} , \qquad (2)$$

and

$$f = \left(\frac{e*}{e} \right)^2 , \qquad (3)$$

where A, τ, E_g, n, and f are the PL intensity component, lifetime(nsec), optical band-gap energy(eV) of GaN corresponding to the PL peak, refraction index, and oscillator strength as the square ratio of charge, respectively. The lifetime constants were very proportional to emission wavelengths for the square well structure and the lifetime for a intense 479-nm excitation is very short to the value of 0.170 nsec, indicating a fast recombination.

Figure 3 shows the TRPL spectra measured on the sample of trapezoidal well structure at room temperature with a fixed pump power of 40 mW. The TRPL spectra were measured at two positions of PL peaks at 457 and 479 nm. The lifetimes show the almost same values of 0.540 and 0.580 nsec, which are the same order to the lifetime of 0.530 nsec for the GaN host material and so there is no lifetime dependence on PL peak positions in the trapezoidal well structure. Furthermore, this structure shows the stable PL spectra independent to external biases such as temperature and voltage[10].

Generally, the recombination rate of exciton can be perturbed by the external bound states such as point defect, dislocation and quantum well[11]. The perturbation potential can reduce the density overlapping between electron and hole so that the recombination rate can be decreased. The recombination rate is the inverse lifetime and the lifetime becomes longer for the bound exciton than for the free exciton. With the strong confinement of quantum well, the bound excitonic lifetimes are ranged to 0.210-0.300 nsec for the InGaN SQW widths of 1.7-5 nm [12-15]. Our result also shows a similar value of the lifetime of 0.17 nsec for the square MQW structure. The quantum well width comparable to the effective Bohr radius of Wannier exciton[16] as the electron-hole correlation distance can enhance the charge density overlapping between electron and hole.

Even though the well width in the InGaN/GaN quantum well structure is smaller than the Bohr radius, the strong piezoelectric field can severely distort the hole state of the valence band offset. The interfacial strain between the InGaN quantum well and GaN barrier provides the internal piezoelectric field and the modification of the band information such as gap, hole mass and the valence-

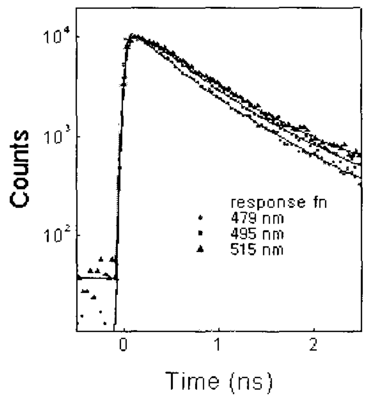

Fig. 2. The room temperature TRPL spectra from the square well structure with a fixed pump power of 40 mW.

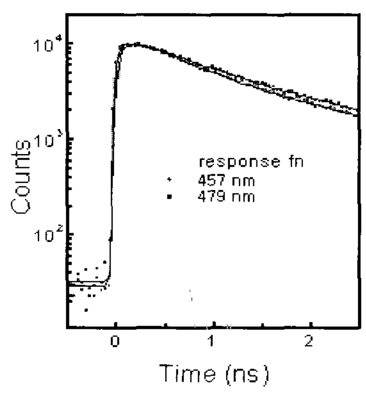

Fig. 3. The room temperature TRPL spectra from the trapezoidal well with a fixed pump power of 40 mW.

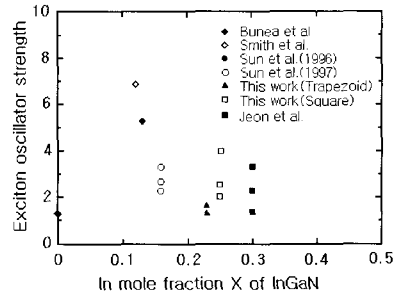

Fig. 4. The oscillator strengths of InGaN/GaN quantum wells for various In mole fractions.

subband structure. This field effect provides the spatial separation between the electron and hole density. This causes the red-shift in momentum space and results in the reduction of the optical band gap energy. The band distortion is more severe in the square well structure rather than trapezoidal well structure.

The oscillator strength is defined by an effective number of oscillators of frequency corresponding to the photon emission energy and can be analyzed by the probability of finding the electron at the position of the hole at a unit cell or a quantum well[9]. Therefore, the faster process with the larger oscillator strength is occurred in a more bound state than the longer process.

Figure 4 shows the oscillator strengths for various In mole fractions of InGaN/GaN QW structures. The strength can be obtained by the analysis of the optical absorption or photon emission from the excited state. As a reference, the oscillator strength for GaN host material is about 1.3[8]. According to Sun et al. [12], as decreasing the well width in the single quantum well structure, the recombination lifetime is decreased corresponding to the increased oscillator strength. The value of oscillator strength is ranged to 2.3-3.3. This indicates that quantum well provides a potential well for both electron and hole and enhances the correlation of between electron and hole densities.

The trapezoidal well structure provides relatively longer lifetime of 0.54 nsec and the relatively small oscillator strength of 1.1 similar to undoped GaN. This indicates that the trapezoidal well does not play a role of the strong confinement in contrast to square well structure. From the rate equation of the combined recombination processes, $1/\tau_r = 1/\tau_{r1} + 1/\tau_{r2}$, the process correlated to the shorter lifetime is more dominantly contributed to the combined transition processes. Therefore, the relatively large value of lifetime for the trapezoidal quantum well implies the less carrier density due to the carrier localization at the interfacial trap state originated from the fluctuation of the In mole fraction or In clustering[17]. This can be verified by the micro-structural investigation of TEM image.

Figure 5 shows the high resolved TEM image of InGaN/GaN MQWs. The square MQW structure shows very distinguishable contrast region between well and barrier. The dark contrast in well region implies the stress concentration. However, for the trapezoidal well structure shows the scattered contrast region in vicinity of the well region and so the well region is not well defined. This unconfined MQW structure provides the weak PL intensity and the un-relaxed structure through the non-uniformed In mole fraction. This In clustering phenomenon in InGaN well region provides the quantum dot-like behavior in the recombination lifetime[17,18].

According to Cho et al.[18], the growth interruption between the InGaN well and GaN barrier in square well MQW structure can enhance the stress relaxation and reduces the In clustering. This results in the decrease the radiative lifetime. Without the interruption, the In clustering effect provides the strained layer in the vicinity of the In cluster region and results in the carrier localization. This low-dimensional PL behavior like the quantum dot structure shows the temperature independent radiative lifetime[17].

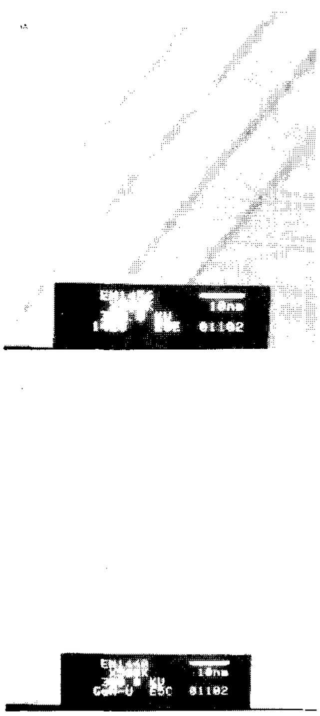

Fig. 5. The micro-structural TEM images of InGaN/GaN multi-quantum wells: (a) square well and (b) trapezoidal well.

4. SUMMARY

In summary, the square- and trapezoidal-type InGaN/GaN multi-quantum wells were investigated for the room temperature time-resolved photoluminescence. The radiative recombination lifetime of 0.170 nsec from square well and 0.540 nsec from trapezoidal well were found at room temperature corresponding to the oscillator strengths of 3 and 1.1, respectively. The relatively long lifetime in trapezoidal well is correlated with the relaxed interface by forming the In clusters. The trapezoidal quantum well with a low PL intensity is strongly associated with the relaxed structure. Therefore, we suggest a method that a highly bright light emission can be achieved by the introduction of the trapezoidal MQW structure grown with the time interruption for the atomic layer epitaxy of the well region.

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