Characterization of high-quality InGaN/GaN multiquantum wells with time-resolved photoluminescence

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Recombination in single quantum well and multiquantum well InGaN/GaN structures is studied using time-resolved photoluminescence and pulsed photoluminescence measurements. Room-temperature measurements show a rapid lifetime (0.06 ns) for a single quantum well structure, while an increasingly long decay lifetime is measured for multiquantum wells as more quantum wells are incorporated into the structure. Temperature-dependent lifetime measurements show that a nonradiative recombination mechanism activates above 45 K in the single quantum well but is less important in the multiquantum wells. © *1998 American Institute of Physics.* [S0003-6951(98)02209-8]

The demonstration of high-brightness light-emitting diodes, and laser diodes with InGaN active regions[1,2] has established the III–V nitrides as an important material system for optoelectronics operating in the green–UV range. Because of the high density of structural and electronic defects present, as well as the strain inherent in growth of these lattice-mismatched materials, characterization of the dominant recombination mechanisms is critical to understand the mechanisms of efficient optical emission. This study carries out systematic measurements of the temperature-dependent lifetimes of single and multiple $In_{0.2}Ga_{0.8}N$ quantum wells. The quantum wells (QW) are nominally identical: the differences in measured lifetimes may provide important insight into critical features of the growth and formation of highly efficient optical structures.

$In_{0.2}Ga_{0.8}N$/GaN quantum wells grown for this study were deposited by metal–organic chemical-vapor deposition on *C*-plane sapphire in an atmosphere pressure reactor. After annealing the substrate at 1050 °C, a 19 nm thick GaN nucleation layer was deposited at 600 °C. The temperature was then raised to 1080 °C to grow GaN:Si (1×10^{18} cm^{-3}) of 1.8 μm thickness. The quantum well structures consisting of 7 nm GaN:Si (1×10^{18} cm^{-3}) barriers and 2 nm $In_{0.2}Ga_{0.8}N$ [nonintentionally doped (nid) *n*-type $3-5\times10^{17}$ cm^{-3}] wells were grown next and capped with 100 nm of $Al_{0.06}Ga_{0.94}N$ (nid *n*-type $3-5\times10^{17}$ cm^{-3}) to prevent surface recombination. The quantum well samples comprised either a single quantum well (SQW), or multiple quantum well (MQW) structures with 5 (5QW) and 10 (10QW) nominally identical $In_{0.2}Ga_{0.8}N$ quantum wells. The samples were grown in rapid succession, ensuring as far as possible, the uniformity of the quantum well structures.

Time-resolved photoluminescence (TRPL) and pulsed photoluminescence (PL) were measured using a tunable Ti: sapphire laser operating at a repetition rate of 80 MHz. The 150 fs pulses from the laser were doubled and focused onto the sample with a spot diameter of 100 μm and a maximum laser power of 120 mW. Temperature-dependent TRPL measurements were taken at two different excitation energies. The laser energy was tuned for excitation above the barrier energy ($E_{exc}=3.47$ eV) and below the barrier energy but above the $n=1$ heavy-hole transition in the wells ($E_{exc}=3.26$ eV). Pulsed PL measurements were taken at an excitation energy of 3.26 eV. Assuming 100% collection efficiency, the injected carrier density was 8×10^{16} cm^{-3} for excitation of the quantum wells only ($E_{exc}=3.26$ eV), and 1×10^{18} cm^{-3} for excitation above the barrier energy ($E_{exc}=3.47$ eV). The photoluminescence was collected and focused onto a half-meter monochromator. The monochromator was scanned to obtain photoluminescence spectra, and set to select photoluminescence at spectral peaks for time-resolved decay measurements. The exit slit of the spectrometer was removed, and a Hamamatsu streak camera placed at the output focal plane of the monochromator. The spectrometer was set at the peak emission wavelength so that the time-resolved data were centered at the peak with a 10 nm bandwidth. The system resolution was ~60 ps.

Figure 1 compares pulsed photoluminescence spectra from the samples at low temperature for the SQW, 5QW, and 10QW samples. The main emission peak is due to re-

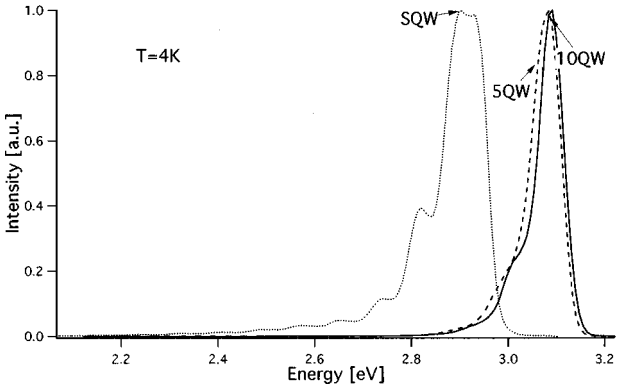

FIG. 1. PL spectra of quantum well structures at $T=4$ K.

TABLE I. Summary of photoluminescence data.

Number of quantum wells	Internal quantum efficiency (%)	RT Pk (eV)	4 K Pk (eV)	RT FWHM (eV)	4 K FWHM (eV)
1	0.37	2.931	2.931	0.111	0.097
5	10.5	3.031	3.085	0.096	0.077
10	31.8	3.061	3.085	0.098	0.058

combination in the $In_{0.2}Ga_{0.8}N$ well region. The 10QW structure shows the narrowest linewidth of 58 meV at low temperature. A calculation of the emission energy at room temperature for the $n=1$ transition in an (unstrained) $In_{0.2}Ga_{0.8}N$ quantum well gives 3.096 eV. Allowing for the effects of strain and compensating piezoelectric fields[3] would redshift this energy by 10 s of meV giving reasonable agreement with the measured emission energy for the MQWs. The SQW emission is considerably redshifted from the MQW photoluminescence peak energies. It is possible that this shift is due to well width variation, a higher degree of strain, or a higher In concentration in the SQW.

Table I summarizes important properties of the photoluminescence data for the SQW and MQW structures. The peak position (Pk) and full width at half maximum (FWHM) at low temperature and at room temperature (RT) are given. Jackson and Yablonovitch[4] measured the internal quantum efficiency (iqe) of these samples at room temperature using a 14 mW continuous-wave HeCd laser at a 325 nm pump wavelength. The iqe is highest for the 10QW structure, indicating high radiative efficiency at room temperature for this sample. The SQW shows very low efficiency suggesting that nonradiative processes are dominating recombination in this structure at room temperature. Optical pumping data from laser structures incorporating varying numbers of quantum wells into the laser active region were also measured by coworkers. Lasing was not observed for a SQW and 5QW structure. The 10QW structure, however, lased when optically pumped.[5]

Time-resolved photoluminescence decay measurements taken with the streak camera at $E_{exc}=3.47$ eV are presented in Figs. 2 and 3. To account for inhomogeneity across the photocathode, the data traces were normalized by the measured cathode sensitivity (shading calibration). This proce-

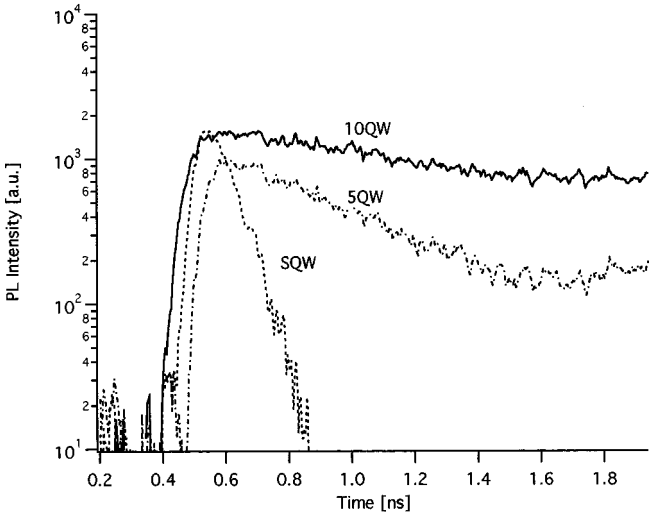

FIG. 2. Room-temperature time-resolved photoluminescence for increasing number of quantum wells.

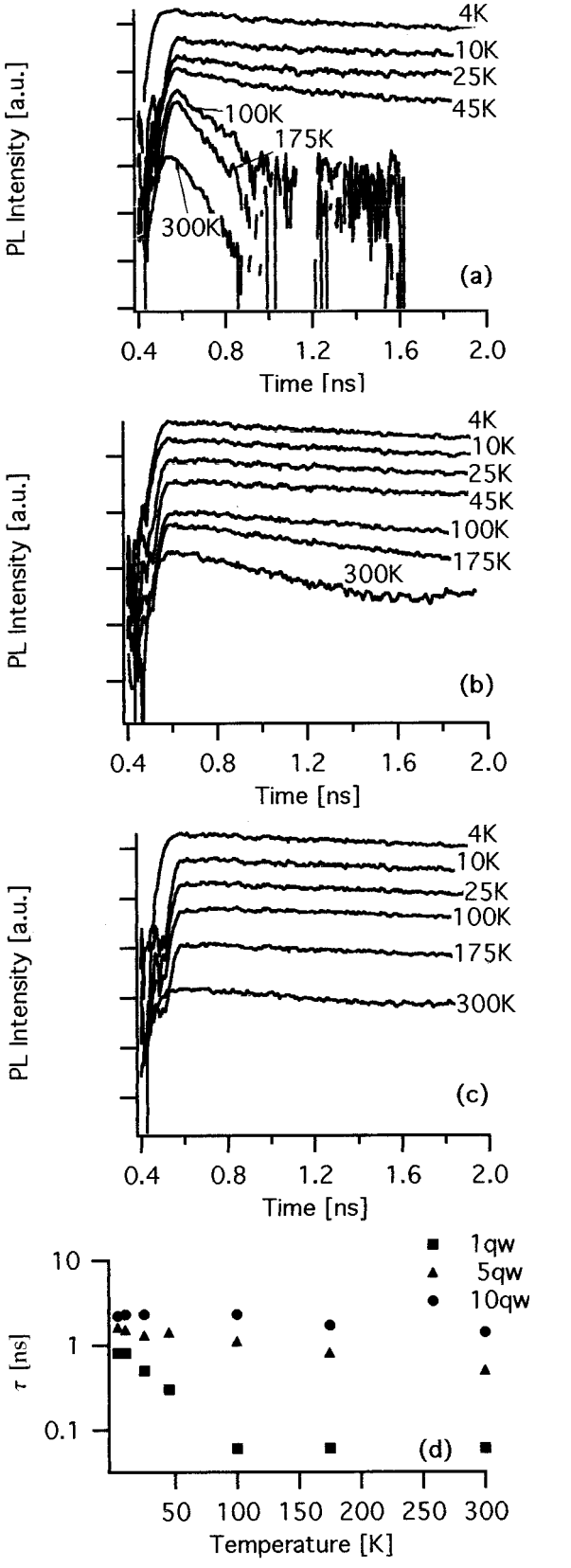

FIG. 3. Time-resolved photoluminescence as a function of temperature (spectra are offset for clarity). (a) SQW; (b) 5 QW; (c) 10 QW; and (d) lifetime vs T.

dure degrades our signal-to-noise ratio and explains correlations in the noise on the various traces. The photoluminescence lifetime was determined by fitting the data with an exponential decay convolved with the system impulse response.

Figure 2 compares room-temperature transient photoluminescence data from the SQW and MQW structures. A convolution fit to the SQW data gives a rapid single exponential decay with $\tau_{\text{meas}} \sim 0.06$ ns, the system resolution. The TRPL spectra for the multiple quantum well samples show an increasingly long lifetime as the number of quantum wells is increased.[6] $\tau_{\text{meas}} = 0.5$ ns for the 5QW sample and 1.4 ns for the 10QW sample. Figure 3 tracks the temperature dependence of τ_{meas} for the different samples. The decay time is a strong function of temperature for the SQW. It begins to decrease sharply above 45 K, decreasing from 0.8 ns at low temperature to 0.06 ns at room temperature. This temperature dependence suggests the activation of nonradiative recombination mechanisms with increasing temperature. The temperature dependence is weaker for the 5 and 10 quantum well samples [Fig. 3(d)].

In addition, $I(T)$ the peak emission intensity as a function of temperature, drops by almost two orders of magnitude from 4 to 300 K for the SQW structure, while remaining relatively insensitive to temperature for the 10QW sample. These results indicate that, at room temperature, the SQW luminescence decay has a stronger nonradiative component than the MQW decays. A possible source of nonradiative recombination in the SQW sample may be interface states. Such a nonradiative component was apparent from our previous work on the growth rate dependence of recombination lifetimes in InGaN/GaN SQWs.[7] Here, our measurements show a clear improvement in luminescence properties with an increasing number of quantum wells. One reason for the improved emission may lie in the strain-induced gettering of impurities as the number of grown interfaces increases. Such factors have been reported for the GaAs/AlGaAs materials system.[8] The photoluminescence decay curves observed would then be a superposition of luminescence from initial well(s) with lower efficiency with subsequent wells of improved quality. The overall efficiency of the structures would also increase with increasing number of wells.

Thermal emission from the wells is probably a less important mechanism for carrier loss than nonradiative recombination at interface states. The 50% point on the low-energy side of the photoluminescence peak was measured for the GaN barriers and found to be 3.38 eV at room temperature. This is 250 meV above the high-energy-side 50% point of the highest-energy quantum well photoluminescence peak observed for this sample set. Assuming a 70/30 conduction/valence-band offset, electrons and holes should be confined by 3–4 kT or more at room temperature in all samples.

Localized carriers have been proposed as a spontaneous emission mechanism[9,10] for InGaN bulk and InGaN/GaN MQW light-emitting diodes and laser diodes. The carriers are believed to be bound by states created due to microcrystalline disorder. The samples used in this study were grown with a nominal In composition of $x = 0.2$. Theoretical studies indicate a bimodal concentration of only 6% at 800 °C, and a large solid immiscibility gap that covers almost the entire range of $In_xGa_{1-x}N$ alloys. This results in almost perfect kinetic phase separation for In concentrations above $x = 0.10$, as observed by Osamura *et al.*[11] Such a separation generally originates from interface states, and studies on GaInP/InP quantum wells indicate that the resulting strain fields propagate through a multiple quantum well structure. The separated phase structure becomes more pronounced with an increasing number of wells, perhaps giving rise to highly efficient radiative centers with a long radiative lifetime.

The combined effect of a reduced density of nonradiative centers and an increase in compositional fluctuation as more wells are deposited would result in the observed increase of both efficiency and lifetime with an increasing number of wells. It should be noted, however, that an increased density of below-band localizing states due to compositional fluctuation would result in more emission at longer wavelengths for the MQWs. This trend is not observed for these samples.

In summary, we have carried out systematic temperature-dependent photoluminescence and photoluminescence decay measurements on nominally identical single and multiple quantum wells. There is strong correlation of both lifetime and luminescence efficiency with material structure. Both quantities increase with an increasing number of quantum wells. The explanation for this correlation may reside either with the role of interfaces in gettering defects through the growth process, or with an increased compositional modulation of the material as the number of quantum wells is increased. Further experiments are needed to better understand this correlation.

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