

Enhanced luminescence in InGaN multiple quantum wells with quaternary AlInGaN barriers

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We report on the comparative photoluminescence studies of AlGaIn/GaN, GaN/InGaIn, and AlInGaIn/InGaIn multiple quantum well (MQW) structures. The study clearly shows the improvement in materials quality with the introduction of indium. Our results point out the localized state emission mechanism for GaN/InGaIn structures and the quantum well emission mechanism for AlInGaIn/InGaIn structures. The introduction of indium is the dominant factor responsible for the observed differences in the photoluminescence spectra of these MQW structures. © 2000 American Institute of Physics. [S0003-6951(00)00743-9]

Built-in electric fields related to strain and spontaneous polarization play a very important role in GaN/AlN/InN-based devices.¹ Recently, we proposed and demonstrated an approach for controlling these fields using abrupt and graded heterointerfaces with quaternary AlInGaIn layers.² This strain and energy band Engineering approach allowed us to adjust strain and band gap offsets independently, and we applied this technique for the design and optimization of heterostructure field effect transistors.³

In this letter, we report on the photoluminescence studies of AlInGaIn/InGaIn multiple quantum well (MQW) structures. The introduction of indium into the barrier layers can affect the MQW properties via three different mechanisms. First, the indium molar fraction might have a very pronounced effect on the overall strain and on the strain in the quantum wells. According to the first order theory of elasticity, thin multiquantum well layers simply adjust to the lattice constants of the buffer layer. In this case, the introduction of indium into the barrier layers would only change strain in these layers, and not in the quantum wells, assuming that the structure is fully strained. However, our early results on GaN/AlN/GaN semiconductor-insulator-semiconductor structures⁴ clearly showed that a partial strain relaxation might occur at the film thickness well below the critical thickness. Therefore, the magnitude of strain in the barrier layers might directly affect the dislocation density and the materials quality of the quantum wells. Second, the magnitude of the built-in fields induced by the spontaneous polarization should be affected. The introduction of quaternary barriers should allow us to independently control both spontaneous polarization and strain. This approach can be called polarization energy band engineering. Finally, our previous results⁵ show that the introduction of indium improves the materials quality and surface morphology. The results of the present study clearly confirm this improvement in materials quality with the introduction of indium and show this to be

the dominant factor responsible for the observed differences in the photoluminescence spectra.

AlInGaIn-GaN MQW structures were grown on n^+ -SiC substrates following a 0.8- μ -thick n^+ -Al_{0.1}Ga_{0.9}N conducting buffer layer and a 0.1- μ m-thick n -GaN layer. All the layers in the structures were grown by low pressure metalorganic chemical vapor deposition at 76 Torr and 1000 °C. However, the growth temperature and pressure for the MQW layers for the structures was kept constant at 780 °C and 200 Torr, respectively. All of the other growth details were the same as reported earlier.^{2,3} The MQW region consisted of two In_{0.2}Ga_{0.8}N quantum wells surrounded by three barrier layers. The barrier layer for the sample was Al_{0.15}In_{0.05}Ga_{0.8}N. The well and barrier layer thicknesses were kept at 30 and 40 Å, respectively. The alloy composition for the quaternary Al_xIn_yGa_{1-x-y}N barrier layer was determined using a separate 2000-Å-thick layer and the procedure outlined in our earlier publications.^{2,3} The growth time was then scaled to achieve the desired barrier layer thickness. In Fig. 1 we include the secondary ion mass spectroscopy (SIMS) profiles for a representative Al_{0.15}In_{0.05}Ga_{0.8}N-In_{0.2}Ga_{0.8}N MQW sample with two wells and three barriers. The quantum well and the buffer layer growth conditions for this sample of Fig. 1 were identical to the three structures of our study. The SIMS data clearly shows a simultaneous presence of Al and In in the barrier regions. It also shows the In peaks corresponding to the two InGaIn wells.

Room-temperature photoluminescence (PL) was then measured for the Al_{0.15}In_{0.05}Ga_{0.8}N barrier MQW sample using a low intensity (10 mW) HeCd laser operating at $\lambda = 325$ nm. For comparison the PL spectra for a GaN-In_{0.2}Ga_{0.8}N MQW sample with identical well and barrier thickness was also measured. These PL spectra are included in Fig. 2. As seen from the data of Fig. 2, the addition of Al and In to the GaN barrier of a GaN-In_{0.2}Ga_{0.8}N MQW results in a strong blueshift of the peak wavelength of the PL signal. It also significantly increases the intensity of the PL emission. There can be several possible mechanisms for this blue shift and increased PL intensity for the quaternary barrier MQW structure. These are: (i) stronger carrier confine-

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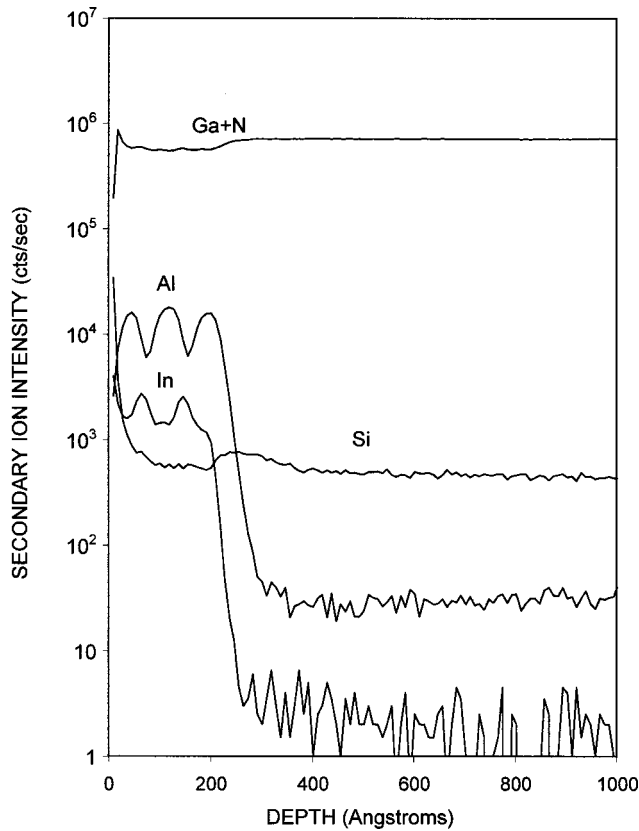


FIG. 1. SIMS profiles of the quaternary $\text{Al}_{0.15}\text{In}_{0.15}\text{Ga}_{0.7}\text{N}$ - $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$ MQW.

ment due to a larger band gap offset resulting from the use of $\text{Al}_{0.15}\text{In}_{0.05}\text{Ga}_{0.8}\text{N}$ barriers; (ii) possible reduction in the polarization charge, and hence, in a built-in field across the quantum well; and finally, (iii) improvement of quantum well quality resulting in a reduction of the band tail states. To determine which of the earlier is the dominant mechanism further experiments were carried out.

We first measured the PL spectra for an $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$ MQW sample with $\text{Al}_{0.15}\text{Ga}_{0.85}\text{N}$ barrier layers. This spectrum is also included in Fig. 2. In spite of the increased confinement we observe a strong redshift and a weaker PL emission signal. This rules out carrier confinement as the

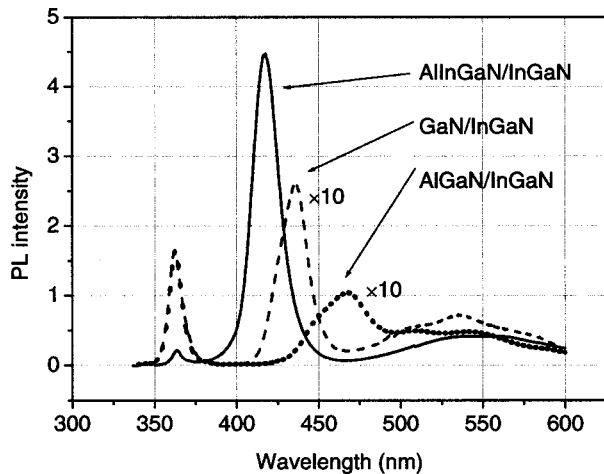


FIG. 2. Room-temperature PL spectrum comparison of $\text{Al}_{0.15}\text{In}_{0.05}\text{Ga}_{0.8}\text{N}$ - $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$, GaN - $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$, and $\text{Al}_{0.15}\text{Ga}_{0.85}\text{N}$ - $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$ MQW.

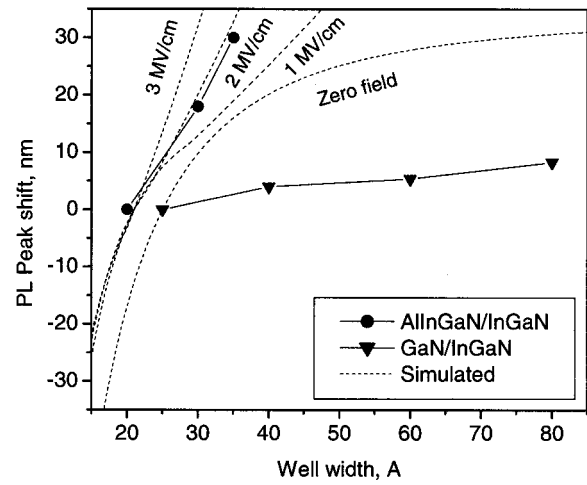


FIG. 3. PL peak shift as a function of the quantum well width. Circle: $\text{Al}_{0.15}\text{In}_{0.05}\text{Ga}_{0.8}\text{N}$ - $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$ MQW, triangle: GaN - $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$ MQW. Dashed lines present the theoretical results for the electrical field across the quantum wells 0...3 MV/cm. To compare with experiments, the offset (zero shift) for the zero-field calculation corresponds to the 25 Å well width, while for the nonzero fields the offsets correspond to 20 Å wide QW.

dominant mechanism for the observed PL emission enhancement and blueshift of our quaternary barrier MQW. **For built-in field to be the dominant mechanism for the blueshift and increased PL emission intensity the PL peak position should be strongly dependent on the width of the quantum wells.**^{6,7} To establish the role of built-in field for our $\text{Al}_{0.15}\text{In}_{0.05}\text{Ga}_{0.8}\text{N}$ - $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$ MQW we fabricated structures with quantum well widths ranging from 20 to 50 Å. The barrier layers were kept fixed at 60 Å. For a comparison similar geometry GaN - $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$ MQW structures were also grown. In Fig. 3 we plot the relative shift of the PL peak position as a function of the quantum well width for the AlInGaN (circles) and the GaN (triangles) barrier MQW structures. Since the absolute position of PL peak differs for QWs with AlInGaN and GaN barriers the shift was calculated with respect to the narrowest QW peak position.

Also plotted in Fig. 3 are the calculated values of these dependences (dashed and dotted lines). The following approach was used to simulate the PL peak position shift. In the presence of built-in field, F , for infinitely deep potential well, the position of the lowest confined electron energy level, E_0 , is given by triangle well model⁸

$$\delta E_0 = \frac{(2.25\pi\hbar qF)^{2/3}}{(8m_e)^{1/3}}, \quad (1)$$

where m_e is the electron effective mass and q is the electronic charge. The width of this quantum state can be then found as

$$d_0 = E_0 / (qF). \quad (2)$$

If the width of the finite QW, d , is larger than d_0 , the position of quantum confined energy level can be estimated using Eq. (1). However, if the QW is narrower than d_0 , the quantum-confined state is closer to the lowest energy level position in rectangular QW of the width d :

$$E_1 = \frac{\pi^2\hbar^2}{2m_e d^2}. \quad (3)$$

The transition from field-defined to the rectangular QW quantization occurs when the width of the triangle well quantum state, d_0 , becomes equal to the QW width, d . From Eqs. (1) and (2), we find the critical built-in field corresponding to this transition

$$F_c = \frac{6.245\hbar^2}{d^3 q m_e}. \quad (4a)$$

If the QW width, d , is expressed in angstroms, then the critical field, F_c , in volts per centimeter, can be easily calculated from (4a) as

$$F_c = 4.8 \times 10^9 / (d^3 m_c / m_0). \quad (4b)$$

Similar expressions can be written for light and heavy hole quantum levels. The shift of PL spectrum with respect to the QW band gap can now be found as

$$\Delta E = \delta E_e + \delta E_h - qFd, \quad (5)$$

where δE_e and δE_h are the displacements of electron and holes confined states calculated from (1) or (3) depending on criterion (4a); the last term in (5) accounts for field-induced redshift of the PL emission peak. Figure 3 (dashed curves) shows the results of our calculations for different values of built-in field across the QW. The values of the effective masses $m_e = 0.23m_0$ and $m_h = 2m_0$ for electrons and for heavy holes, respectively, were used in these calculations. To compare the simulations with the experimental data for AlInGaN/InGaN and GaN/InGaN QWs, the simulated shifts for $F = 1, 2$, and 3 MV/cm are offset by the shift corresponding to 20 Å QW width, while the shift for zero field is offset by the shift corresponding to 25 Å wide well.

Our experimental data for the GaN-In_{0.2}Ga_{0.8}N show very weak dependence on the QW width. As seen from the simulated dependencies, even for zero built-in field the significant PL peak position dependence on QW width for narrow wells (below 30 Å) has to be observed due to quantum confinement effects. Besides, it is well known^{1,6} that a strong built-in field is always present in GaN-InGaN quantum wells due to significant lattice mismatch. Hence, the dominant mechanism for the PL emission in our GaN-InGaN MQW cannot be explained by pure QW emission. Conversely, the observed shifts agree very well with the mechanism of localized states (quantum dots) emission.

The quaternary barrier structures demonstrate a different mechanism of the PL emission. For the AlInGaN-InGaN structure the measured PL peak position shift agrees well with calculated values taking the built in field at 2 MV/cm. This value is very close to the polarization field estimated for our structures assuming no relaxation in the barrier layer and quantum well. Thus, we conclude that the PL emission for our Al_{0.15}In_{0.05}Ga_{0.8}N-In_{0.2}Ga_{0.8}N structure behaves more like that from a conventional quantum well rather than from localized states.

To further confirm this assessment, we measured the PL emission from the quaternary AlInGaN and the GaN barrier InGaN MQWs as a function of optical excitation level. The increased carrier concentration at higher excitation levels can give rise to: (i) band filling for which width independent

blueshift should occur; (ii) band renormalization for which width independent redshift should occur; and (iii) built-in field screening where the blueshift is width dependent. We then measured the maximum relative shift of the PL peak position $\Delta\lambda$ ($\Delta\lambda = \text{PL peak position at low intensity} - \text{PL peak position at high intensity}$) as a function of quantum well width for the GaN-InGaN and the AlInGaN-InGaN MQW structures. For GaN-InGaN MQW the maximum blueshift depends weakly on the QW width. However for the AlInGaN-InGaN MQW well-pronounced field screening is clearly visible. This again confirms the localized state emission mechanism for GaN/InGaN and QW emission mechanism for AlInGaN/InGaN structures.

For the Al_{0.15}In_{0.05}Ga_{0.8}N-In_{0.2}Ga_{0.8}N MQW sample PL spectra were measured for temperatures ranging from 6.5 to 300 K. These data show a blueshift of the peak emission wavelength with decreasing temperatures, which we attribute to an increase of the band gap energy. From the temperature dependence of PL intensity, we clearly observe an activation behavior with the activation energy of 30 meV. This agrees well with estimated binding energy value of 30.4 meV for two-dimensional confined excitons in GaN.^{9,10} Thus again, the luminescence mechanism of our quaternary quantum wells appears to be free excitonic recombination rather than that from bound excitons localized at the potential minima.

In summary, we have presented a study of luminescence properties of InGaN MQWs with quaternary AlInGaN barriers. Our results demonstrate that quaternary barriers (with InGaN quantum wells) change the PL emission mechanism from localized state/quantum dots based to that of a conventional quantum well structure. We attribute this change to a drastic improvement in the heterostructure quality resulting from the use of quaternary barrier layers. This approach should enable the design of more efficient solid-state light emitters.

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¹T. Takeuchi, C. Wetzel, S. Yamaguchi, H. Sakai, H. Amano, I. Akasaki, Y. Kaneko, S. Nakagawa, Y. Yamaoka, and N. Yamada, Appl. Phys. Lett. **73**, 1691 (1998).

M. Asif Khan, J. W. Yang, G. Simin, R. Gaska, and M. S. Shur, Appl. Phys. Lett. **76**, 1161 (2000).

M. Asif Khan, J. W. Yang, G. Simin, R. Gaska, M. S. Shur, and A. Bykhovskiy, Appl. Phys. Lett. **75**, 2806 (1999).

⁴A. Bykhovskiy, B. Gelmont, and M. Shur, J. Appl. Phys. **74**, 6734 (1993).

⁵G. Tamulaitis, K. Kazlauskas, S. Juršėnas, and A. Žukauskas, M. A. Khan, J. W. Yang, J. Zhang, G. Simin, R. Gaska, and M. S. Shur, Appl. Phys. Lett. (to be published).

⁶P. Riblet, H. Hirayama, A. Kinoshita, A. Hirata, T. Sugano, and Y. Aoyagi, Appl. Phys. Lett. **75**, 2241 (1999).

⁷S.-H. Park and S.-L. Chuang, Appl. Phys. Lett. **76**, 1981 (2000).

⁸C. Weisbuch and B. Vinter, *Quantum Semiconductor Structures* (Academic, London, 1991).

⁹P. Ramvall, P. Riblet, S. Nomura, and Y. Aoyagi, J. Appl. Phys. **87**, 3883 (2000).

¹⁰K. P. O'Donnell, R. W. Martin, and P. G. Middleton, Phys. Rev. Lett. **82**, 237 (1999).