Collective effects of interface roughness and alloy disorder in In x Ga 1x N/GaN multiple quantum wells

K. C. Zeng, M. Smith, J. Y. Lin, and H. X. Jiang

Citation: Applied Physics Letters **73**, 1724 (1998); doi: 10.1063/1.122258
View online: http://dx.doi.org/10.1063/1.122258
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/73/12?ver=pdfcov
Published by the AIP Publishing

Collective effects of interface roughness and alloy disorder in $In_xGa_{1-x}N$/GaN multiple quantum wells

K. C. Zeng, M. Smith, J. Y. Lin, and H. X. Jiang[a)]
Department of Physics, Kansas State University, Manhattan, Kansas 66506-2601

(Received 18 May 1998; accepted for publication 9 July 1998)

The collective effects of alloy disorder and interface roughness on optical properties of $In_xGa_{1-x}N$/GaN multiple quantum wells (MQWs) have been studied. The results are compared with those of GaN/AlGaN MQWs and InGaN epilayers. $In_xGa_{1-x}N$/GaN MQWs emit a broad and asymmetrical photoluminescence (PL) band, while GaN/AlGaN MQWs and InGaN epilayers emit narrower and Gaussian-shaped PL bands. Furthermore, the decay of excitons at low temperatures in $In_xGa_{1-x}N$/GaN MQWs follows a nonexponential function even at the lower-energy side of the PL spectral peak, while those in GaN/AlGaN MQWs and in InGaN epilayers follow a single exponential function. Both alloy disorder and interface roughness have to be included in order to interpret the PL emission spectrum and the decay dynamics in $In_xGa_{1-x}N$/GaN MQWs. Important parameters of the $In_xGa_{1-x}N$/GaN MQWs, σ_x, σ_L, and $d\tau/dL$, denoting the alloy disorder, the interface roughness, and the rate of changing of the exciton decay lifetime with well width, respectively, have been deduced. The method developed here can be used to determine σ_x, σ_L, and $d\tau/dL$ in any MQW systems with wells being alloy materials. © *1998 American Institute of Physics.* [S0003-6951(98)03336-1]

The group III-nitride wide-band-gap semiconductors have attracted much attention recently due to their many important applications, such as blue/UV light-emitting diodes (LEDs), laser diodes (LDs), and high-temperature/high-power electronic devices.[1] InGaN multiple quantum wells (MQWs) are being used as an active medium for commercial high-brightness blue/green LEDs.[2] Continuous-wave (cw) operation of blue LDs at room temperature based on $In_xGa_{1-x}N$/GaN MQWs have also been achieved recently.[3] However, many fundamentally important issues remain to be addressed and understood before we can take full advantage of these materials. One of the important issues is related to the optical properties of the InGaN active layers in $In_xGa_{1-x}N$/GaN MQWs.[4] In conventional III–V MQWs, such as GaAs/AlGaAs MQWs, the well regions are formed by a binary semiconductor. It is fully established that the well width fluctuation or interface roughness is very important in GaAs/AlGaAs MQWs.[5–9] On the other hand, the alloy disorder also strongly influences the optical properties of nitride alloys.[10–12] In $In_xGa_{1-x}N$/GaN MQWs, the well regions are formed by the InGaN alloy, thus both alloy disorder and interface roughness are involved and correlated. When both types of disorders are simultaneously present in the same system, direct methods for determining these parameters do not currently exist.

In this work, we have investigated the collective effects of alloy disorder and interface roughness on the optical properties of $In_xGa_{1-x}N$/GaN MQWs.

The $In_xGa_{1-x}N$/GaN MQW ($x\approx0.15$) sample used here was composed of 45 periods of alternating 25 Å $In_xGa_{1-x}N$ wells and 25 Å GaN barriers, and deposited on a sapphire (0001) substrate by low-pressure metalorganic chemical vapor deposition. Schematic diagrams of the MQW structure with interface roughness and alloy disorder in the $In_xGa_{1-x}N$ well regions are shown in Fig. 1. Detailed information about the time-resolved PL laser spectroscopy system can be found elsewhere.[10]

In Fig. 2(a), we show the cw PL spectrum of the $In_xGa_{1-x}N$/GaN MQW sample measured at 10 K. For comparison, we have also plotted the cw PL spectra for an $In_xGa_{1-x}N$ ($x\approx0.17$) epilayer and a 20 Å/50 Å GaN/$Al_xGa_{1-x}N$ MQWs ($x\approx0.07$) measured at 10 K in Fig. 2(b). Optical transitions observed in these materials at low temperatures are due to the localized exciton recombination, caused either by alloy disorder or interface roughness.[10–13] The PL spectra line shapes for the $In_xGa_{1-x}N$ epilayer and GaN/AlGaN MQWs are of Gaussian shape. This is clearly illustrated by the solid fitting lines in Fig. 2(b). However, for the 25 Å $In_xGa_{1-x}N$/GaN MQWs, a Gaussian function does not fit the spectrum well, due to the asymmetrical line shape.

For $In_xGa_{1-x}N$/GaN MQWs, we denote x_0 as the average In composition, σ_x as the compositional fluctuation parameter, L_0 as the average well width, and σ_L as the interface roughness parameter. The formation of both alloy disorder and interface roughness are random processes, we thus assume that the probability for a local point to have In composition x and well thickness L is

$$P_{x,L}\propto\exp[-(x-x_0)^2/2\sigma_x^2]\exp[-(L-L_0)^2/2\sigma_L^2]. \quad (1)$$

The exciton energy as a function of x and L in the MQWs can be written as

$$E(x,L)=E_g(x)+E_c(L)+E_b, \quad (2)$$

where $E_g(x)$, $E_c(L)$, and E_b are the energy gap of the $In_xGa_{1-x}N$ well, confinement, and binding energy of the exciton, respectively. $E_g(x)$ is expressed as $[\alpha x+\beta(1-x)]$, with α and β being the energy gaps of InN and GaN, respec-

[a)]Electronic mail: jiang@phys.ksu.edu

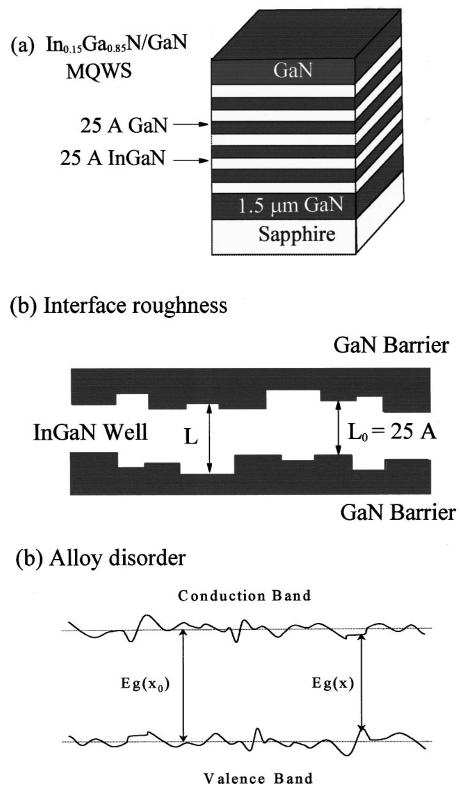

FIG. 1. Schematic diagrams of (a) $In_xGa_{1-x}N$/GaN MQWs, (b) interface roughness, and (c) energy band-gap fluctuations due to alloy disorder in $In_xGa_{1-x}N$ wells.

tively. For simplicity we have neglected the parabolic dependence of $E_g(x)$ on x. The PL intensity at a fixed energy E due to the localized exciton recombination in the $In_xGa_{1-x}N$/GaN can be calculated from

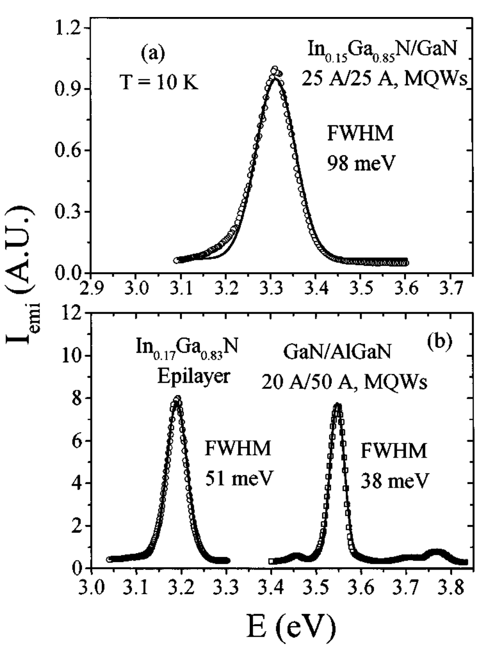

FIG. 2. PL spectra measured at 10 K for (a) 25 Å/25 Å $In_xGa_{1-x}N$/GaN ($x\approx0.15$) MQWs; (b) an $In_xGa_{1-x}N$ ($x\approx0.17$) epilayer (open dots) and a 20 Å/50 Å GaN/$Al_xGa_{1-x}N$ ($x\approx0.07$) MQW sample (open squares). The solid curves are fittings with a Gaussian function. The fitted full widths at half maxima of the PL spectra are 98, 51, and 38 meV for the InGaN/GaN MQWs, InGaN epilayer, and GaN/AlGaN MQWs, respectively.

$$I(E)\propto\int_0^\infty \exp[-(L-L_0)^2/2\sigma_L^2] \times\exp[-(x-x_0)^2/2\sigma_x^2]dL, \quad (3)$$

where $(x-x_0)$ can be written in terms of $(L-L_0)$ and $(E-E_0)$ by using Eq. (2). Here E_0 is the PL peak energy, which corresponds to the energy of an exciton recombined at a site with well width L_0 and In composition x_0. For simplicity, we assume that $E_c(L)$ is proportional to $1/L^2$ and E_b is independent of L and x. Using an approximation to the first order, we get from Eq. (3)

$$I(E)\propto\exp[-(E-E_0)^2/2\sigma_1^2]. \quad (4)$$

Here, $\sigma_1=\Delta E_g[\sigma_x^2+A^2(\sigma_L/L_0)^2]^{1/2}$ with $A=2E_c(L_0)/\Delta E_g$ and $\Delta E_g=(\beta-\alpha)$. Equation (4) thus gives a Gaussian-shaped PL spectrum with a full width at half maximum of $2\sigma_1(2\ln 2)^{1/2}$. The asymmetrical line shape observed for the $In_xGa_{1-x}N$/GaN MQWs shown in Fig. 2(a) points to the fact that higher-order approximation has to be included. If we expand $(x-x_0)$ to the second order of $(L-L_0)$ by using Eq. (2), we get from Eq. (3):

$$I(E)\propto\exp[-(E-E_0)^2/2\sigma_1^2]\exp[-(E-E_0)^3/2\sigma_2^3], \quad (5)$$

where $\sigma_2=\Delta E_g[\sigma_x^{2/3}(\sigma_x^2L_0^2+A^2\sigma_L^2)^{2/3}/(3A^3\sigma_L^4)^{1/3}]$. Equation (5) shows that the PL spectrum of the $In_xGa_{1-x}N$/GaN MQWs is asymmetrical. It has a lower-energy tail, since the higher-order term $\exp(-(E-E_0)^3/2\sigma_2^3)$ favors lower energies. We see that both σ_x and σ_L contribute to the linewidth through the expressions of σ_1 and σ_2 as expected. In Fig. 3, we plotted the PL spectrum of the $In_xGa_{1-x}N$/GaN MQWs together with the fitting by using (a) a Gaussian function and (b) Eq. (5), respectively. The results shown in Fig. 3 demonstrate that Eq. (5) fits the spectrum much better than a Gaussian function. The fitted values of σ_1 and σ_2 are 42 and 88 meV, respectively, from which values of σ_x and σ_L can then be deduced separately. By using 3.503 eV for the band gap of GaN, $\Delta E_g=1.55$ eV, $0.19m_e$ for the electron effective mass, $0.34m_e$ for the hole effective mass, and 67% (33%) for the conduction- (valence-) band offset,[14] we get σ_x and σ_L to be about 0.02 and 3.8 Å, respectively. Our results indicate that the interface quality of the InGaN/GaN MQWs used here is quite good. However, σ_x is larger than a typical value of about 0.01 in II–VI semiconductor alloys,[15] reflecting that it is more difficult to grow $In_xGa_{1-x}N$ alloys.

In Fig. 4, we plotted the PL temporal responses measured at the spectral peak and at a point on the lower-energy side of the spectral peak for the $In_xGa_{1-x}N$/GaN MQWs at 10 K. For comparison, in the inset of Fig. 4, we plotted the 10 K temporal responses measured at the PL spectral peaks for the $In_xGa_{1-x}N$ epilayer and the GAN/AlGaN MQWs. The decay of excitons in the $In_xGa_{1-x}N$ epilayer and GaN/ALGAN MQWs follows a single exponential function. In contrast, the decay of excitons at the spectral peak or at the lower-energy side of the PL spectral peak for the $In_xGa_{1-x}N$/GaN MQWs is nonexponential. In semiconductor alloys (pure alloy disorder) or MQWs with only interface roughness, although nonexponential decay has been observed at the higher-energy side of the PL spectra peak due to the processes of exciton transfer or localization, a single

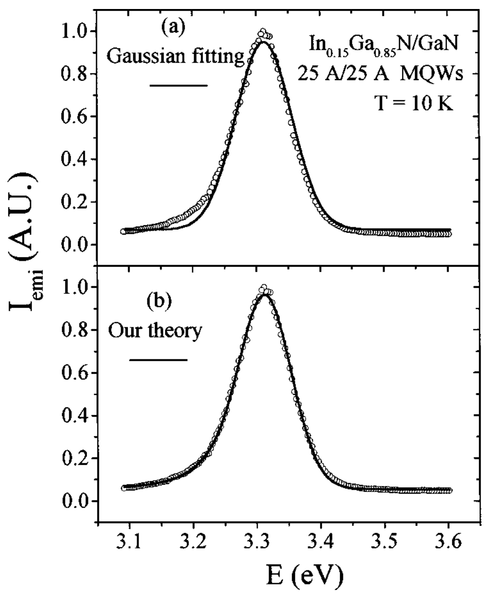

FIG. 3. PL spectrum of the 25 Å/25 Å $In_xGa_{1-x}N/GaN$ MQWs measured at 10 K together with (a) Gaussian fitting; and (b) fitting by Eq. (5). The fitted parameters σ_1 and σ_2 are 42 and 88 meV, respectively.

exponential decay is expected and has been observed at the lower-energy side of the PL spectral peak.[10,13,15]

The PL emission intensity as a function of decay time t can be written as

$$I(E,t) \propto \int_0^\infty \exp[-(L-L_0)^2/2\sigma_L^2] \times \exp[-(x-x_0)^2/2\sigma_x^2]\exp[-t/\tau(L)]dL. \quad (6)$$

In writing Eq. (6), we assume that the exciton lifetime $\tau(L)$ depends only on L, since the dependence of τ on x is much weaker. Expanding $(x-x_0)$ in terms of $(L-L_0)$, $\tau(L)$ as $[\tau(L_0)+(d\tau/dL)|_{L=L0}(L-L_0)]$, we get

$$I(E,t) \propto I(E,0)\exp(-t/\tau_1 + t^2/\tau_2^2), \quad (t<2\tau_2/\tau_1), \quad (7)$$

where $\tau_1 = \tau(L_0)^2(\sigma_x^2 L_0^2 + \sigma_L^2 A^2)/[\tau(L_0)(\sigma_x^2 L_0^2 + \sigma_L^2 A^2) - (d\tau/dL)|_{L=L0} A \sigma_L^2 L_0 (E-E_0)/\Delta E_g]$, $\tau_2 = 2^{1/2}\tau^2(L_0) \times (\sigma_x^2 L_0^2 + \sigma_L^2 A^2)^{1/2}/[\sigma_L \sigma_x L_0 (d\tau/dL)|_{L=L_0}]$, where $\tau(L_0)$ is the exciton decay lifetime at the mean well width L_0. Equation (7), which holds under the condition $t<2\tau_2/\tau_1$, has been used to fit the PL decays of the $In_xGa_{1-x}N/GaN$ MQWs. As demonstrated in Fig. 4, Eq. (7) describes the decay of excitons in $In_xGa_{1-x}N/GaN$ MQWs quite well. The fitted values of τ_1 and τ_2, are 0.75 and 3.6 ns at 3.200 eV, or 0.62 and 3.33 ns at 3.312 eV, respectively.

Furthermore, from the values of τ_1 (=0.62 ns), τ_2 (=3.33 ns), σ_x (=0.02), and σ_L (=3.8 Å), we get $d\tau/dL$ to be about 0.018 ns/Å at the spectral peak for the $In_xGa_{1-x}N/GaN$ MQWs. The value of $d\tau/dL$ obtained here is comparable to a value of 0.013 ns/Å for the GaAs/AlGaAs MQWs.[5] Equation (7) also shows that the PL decay is a single exponential when either σ_L or σ_x is zero, which is what we have observed in Fig. 4 for GaN/AlGaN MQWs and InGaN epilayers.

In conclusion, experimental results reveal (i) a broader and asymmetrical PL spectrum in the $In_xGa_{1-x}N/GaN$ MQWs as compared with the narrower and Gaussian-shaped PL spectra of the GaN/AlGaN MQWs and InGaN epilayers, and (ii) the nonexponential decay of the excitonic transition at the spectral peak or at the lower-energy side of the PL spectral peak in $In_xGa_{1-x}N/GaN$ MQWs as compared with the single exponential decays of the excitonic transitions in the corresponding emission energy regions in the GaN/AlGaN MQWs and InGaN epilayers. These phenomena are due to the collective effects of alloy disorder and interface roughness. $d\tau/dL$ is found to be about 0.018 ns/Å. σ_x and σ_L have been obtained.

This work is supported by DOE (96ER45604), ARO, BMDO/ONR (monitored by Dr. John Zavada and Dr. Yoon S. Park), NSF (DMR-95-28226), and (INT-97-29582).

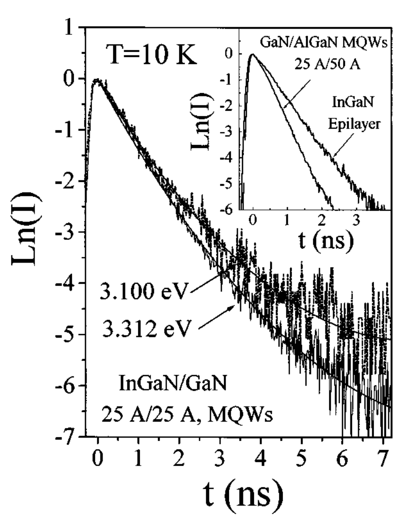

FIG. 4. The main figure shows the PL temporal responses measured at the spectral peak position E=3.212 eV and at a lower-energy E=3.100 eV for the $In_xGa_{1-x}N/GaN$ MQWs measured at 10 K. The inset shows those of the $In_xGa_{1-x}N$ epilayer and $GaN/Al_xGa_{1-x}N$ MQWs measured at the corresponding spectral peaks at 10 K. The solid lines in the main figure are the least-squares fitting in the $In_xGa_{1-x}N/GaN$ MQWs PL decay data with Eq. (7).

[1] H. Morkoç, S. Strite, G. B. Gao, M. E. Lin, B. Sverdlov, and M. Burns, J. Appl. Phys. **76**, 1363 (1994).
[2] S. Nakamura, M. Senoh, N. Iwasa, and S. Nagahama, Jpn. J. Appl. Phys., Part 2 Part 2 **34**, L797 (1995).
[3] S. Nakamura, M. Senoh, S. Nagahama, N. Iwasa, T. Yamada, T. Matsushita, H. Kiyoku, Y. Sugimoto, T. Kozaki, H. Umenoto, M. Sano, and K. Chocho, Appl. Phys. Lett. **72**, 1939 (1998).
[4] S. Nakamura and G. Fasol, *The Blue Laser Diode* (Springer, Berlin, 1997).
[5] J. Feldmann, G. Peter, and E. O. Göbel, Phys. Rev. Lett. **59**, 2337 (1987).
[6] T. Takagahara and E. Hanamum, Phys. Rev. Lett. **56**, 2533 (1986).
[7] P. Zhou, H. X. Jiang, R. Bannwart, S. A. Solin, and G. Bai, Phys. Rev. B **40**, 11 862 (1989).
[8] H. X. Jiang, E. X. Ping, P. Zhou, and J. Y. Lin, Phys. Rev. B **41**, 12 949 (1990).
[9] E. X. Ping and H. X. Jiang, Phys. Rev. B **40**, 11 792 (1989).
[10] M. Smith, G. D. Chen, J. Y. Lin, H. X. Jiang, M. Asif Khan, and Q. Chen, Appl. Phys. Lett. **69**, 2837 (1996).
[11] E. S. Jeon, V. Kozlov, Y. K. Song, A. Vertikov, M. Kuball, A. V. Nurmikko, H. Liu, C. Chen, R. S. Kern, C. P. Kuo, and M. G. Craford, Appl. Phys. Lett. **69**, 4194 (1996).
[12] Y. Narukawa, Y. Kawakami, M. Funato, S. Fujita, and S. Nakamura, Appl. Phys. Lett. **70**, 981 (1997).
[13] M. Smith, J. Y. Lin, H. X. Jiang, A. Salvador, A. Botchkarev, W. Kim, and H. Morkoç, Appl. Phys. Lett. **69**, 2453 (1996).
[14] A. Salvador, G. Liu, W. Kim, O. Aktas, A. Botchkarev, and H. Morkoç, Appl. Phys. Lett. **67**, 3322 (1995).
[15] H. X. Jiang, L. Q. Zu, and J. Y. Lin, Phys. Rev. B **42**, 7284 (1990).