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

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

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The collective effects of alloy disorder and interface roughness on optical properties of In$_x$Ga$_{1-x}$N/GaN multiple quantum wells (MQWs) have been studied. The results are compared with those of GaN/AlGaN MQWs and InGaN epilayers. In$_x$Ga$_{1-x}$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$_x$Ga$_{1-x}$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$_x$Ga$_{1-x}$N/GaN MQWs. Important parameters of the In$_x$Ga$_{1-x}$N/GaN MQWs, $\sigma_x$, $\sigma_L$, 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 $\sigma_x$, $\sigma_L$, and $d\tau/dL$ in any MQW systems with wells being alloy materials. © 1998 American Institute of Physics.

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. InGaN multiple quantum wells (MQWs) are being used as an active medium for commercial high-brightness blue/green LEDs. Continuous-wave (cw) operation of blue LDs at room temperature based on In$_x$Ga$_{1-x}$N/GaN MQWs have also been achieved recently.

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$_x$Ga$_{1-x}$N/GaN MQWs. 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. On the other hand, the alloy disorder also strongly influences the optical properties of nitride alloys. In In$_x$Ga$_{1-x}$N/GaN MQWs, the well regions are formed by the InGaN layer, 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$_x$Ga$_{1-x}$N/GaN MQWs.

The In$_x$Ga$_{1-x}$N/GaN MQW ($x=0.15$) sample used here was composed of 45 periods of alternating 25 Å In$_x$Ga$_{1-x}$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$_x$Ga$_{1-x}$N well regions are shown in Fig. 1. Detailed information about the time-resolved PL laser spectroscopy system can be found elsewhere.

In Fig. 2(a), we show the cw PL spectrum of the In$_x$Ga$_{1-x}$N/GaN MQW sample measured at 10 K. For comparison, we have also plotted the cw PL spectra for an In$_x$Ga$_{1-x}$N ($x=0.17$) epilayer and a 20 Å GaN/Al$_x$Ga$_{1-x}$N MQWs ($x=0.07$) 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.

The PL spectra line shapes for the In$_x$Ga$_{1-x}$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$_x$Ga$_{1-x}$N/GaN MQWs, a Gaussian function does not fit the spectrum well, due to the asymmetrical line shape.

For In$_x$Ga$_{1-x}$N/GaN MQWs, we denote $x_0$ as the average In composition, $\sigma_x$ as the compositional fluctuation parameter, $L_0$ as the average well width, and $\sigma_L$ 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

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

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

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

where $E_g(x)$, $E_c(L)$, and $E_b$ are the energy gap of the In$_x$Ga$_{1-x}$N well, confinement, and binding energy of the exciton, respectively. $E_g(x)$ is expressed as $\left[ ax + \beta \left( 1 - x \right) \right]$, with $a$ and $\beta$ being the energy gaps of InN and GaN, respectively.
due to the localized exciton recombination in the

\[ E_g \]

FIG. 2. PL spectra measured at 10 K for 

different samples. For simplicity, we have neglected the parabolic dependence of \( E_s(x) \) on \( x \). The PL intensity at a fixed energy \( E \) due to the localized exciton recombination in the InGa\(_{1-x}\)N/GaN can be calculated from

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

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 recombinated at a site with well width \( L_0 \) and In composition \( x_0 \). For simplicity, we assume that \( E_s(L) \) 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)

\[ I(E) \approx \exp[-(E-E_0)^2/2\sigma_0^2]. \]

Here, \( \sigma_1 = \Delta E_s[\sigma_x^2 + A^2(L/L_0)^2]^{1/2} \) with \( A = 2E_s(L_0)/\Delta E_s \) and \( \Delta E_s = (\beta - \alpha) \). 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 InGa\(_{1-x}\)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):

\[ I(E) \approx \exp[-(E-E_0)^2/2\sigma_0^2]\exp[-(E-E_0)^2/2\sigma_1^2], \]

where \( \sigma_2 = \Delta E_s[\sigma_x^2 + A^2(L/L_0)^2]^{1/2}/(3A^3 \sigma_L^4)^{1/3} \). Equation (5) shows that the PL spectrum of the InGa\(_{1-x}\)N/GaN MQWs is asymmetrical. It has a lower-energy tail, since the higher-order term \( \exp(-(E-E_0)^2/2\sigma_1^2) \) favors lower energies. We see that both \( \sigma_0 \) and \( \sigma_1 \) contribute to the linewidth through the expressions of \( \sigma_1 \) and \( \sigma_2 \) as expected. In Fig. 3, we plotted the PL spectrum of the InGa\(_{1-x}\)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 \( \sigma_1 \) and \( \sigma_2 \) are 42 and 88 meV, respectively, from which values of \( \sigma_0 \) and \( \sigma_2 \) can then be deduced separately. By using 3.503 eV for the band gap of GaN, \( \Delta E_s = 1.55 \) eV, 0.19\( m_e \) for the electron effective mass, 0.34\( m_0 \) for the hole effective mass, and 67\% (33\%) for the conduction- (valence-) band offset,\(^{14} \) we get \( \sigma_1 \) and \( \sigma_2 \) to be about 0.02 and 3.8 Å, respectively. Our results indicate that the interface quality of the InGaGaN MQWs used here is quite good. However, \( \sigma_x \) is larger than a typical value of about 0.01 in II–VI semiconductor alloys,\(^{15} \) reflecting that it is more difficult to grow InGa\(_{1-x}\)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 InGa\(_{1-x}\)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 InGa\(_{1-x}\)N epilayer and the GaN/AlGaN MQWs. The decay of excitons in the InGa\(_{1-x}\)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 InGa\(_{1-x}\)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
exponential decay is expected and has been observed at the lower-energy side of the PL spectral peak.\textsuperscript{10,13,15}

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

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

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

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

where $\tau_1=\tau(L_0)^2\left(\sigma^2 L_0^2 + \sigma_x^2 A^2\right)/\left[\tau(L_0)^2 + \sigma_x^2 A^2\right] - \left(\tau(L)\right)\left[\tau(L_0)^2 + \sigma^2 A^2\right] - \left(t d\tau/dL\right)\left[\tau(L_0)^2 + \sigma^2 A^2\right] \times (\sigma^2 L_0^2 + \sigma_x^2 A^2)^{-1/2}\left[\sigma \tau(L_0) + t d\tau/dL\right]_{L=L_0} \times \left[\sigma^2 L_0^2 + \sigma_x^2 A^2\right]^{1/2},$ $\tau_2 = 2^{1/2} \tau(L_0) \times (\sigma^2 L_0^2 + \sigma_x^2 A^2)^{-1/2}[\sigma \tau(L_0) + t d\tau/dL]_{L=L_0}$, 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$_{0.25}$Ga$_{1-x}$/N/GaN MQWs. As demonstrated in Fig. 4, Eq. (7) describes the decay of excitons in In$_{0.25}$Ga$_{1-x}$/N/GaN MQWs quite well. The fitted values of $\tau_1$ and $\tau_2$ 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 $\tau_1$ (=0.62 ns), $\tau_2$ (=3.33 ns), $\sigma_x$ =0.02, and $\sigma_L$ =3.8 Å, we get $d\tau/dL$ to be about 0.018 nsÅ at the spectral peak for the In$_{0.25}$Ga$_{1-x}$/N/GaN MQWs. The value of $d\tau/dL$ obtained here is comparable to a value of 0.013 nsÅ for the GaAs/AlGaAs MQWs.\textsuperscript{5} Equation (7) also shows that the PL decay is a single exponential when either $\sigma_1$ or $\sigma_x$ 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$_{0.25}$Ga$_{1-x}$/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$_{0.25}$Ga$_{1-x}$/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 nsÅ. $\sigma_x$ and $\sigma_L$ have been obtained.

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