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Luminescence dynamics and structural investigation of InGaN/GaN multiple quantum well light emitting diodes

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Abstract. Luminescence properties of blue emission InGaN/GaN multiple quantum well (MQW) have been studied by temperature dependent photoluminescence (PL), photoluminescence excitation (PLE) and time-resolved photoluminescence (TRPL) spectroscopic techniques. Two typical samples are studied, both consisting of five periods of InGaN wells with different indium compositions of 21% and 24%, respectively. According to the PL and PLE measurement results, large values of activation energy and Stokes' shift are obtained. This indicates that higher Indium composition results in an increase of composition fluctuation in the InGaN MQW region, showing the stronger carrier localization effect. The lifetime at the low-energy side of the InGaN peaks is longer for higher indium composition, as expected from the larger Stokes shift.

Introduction

Recently, InGaN/GaN multiple quantum wells (MQWs) have attracted much research interest, acting as the active layer in high brightness III-Nitride LED and continuous wave (CW) blue-green laser diode [1-4]. InGaN/GaN heterostructures can exhibit intense photoluminescence (PL) despite a high density of dislocations and defects. More research efforts have been made on InGaN/GaN MQW structures. Most InGaN/GaN MQWs were prepared by MOCVD which has proved to be a powerful technology for industry production of III-nitride materials and devices. In this paper, we are to compare the optical and structural properties with different indium compositions for an InGaN/GaN five-period quantum well structure. Optical characterizations include photoluminescence (PL), photoluminescence excitation (PLE), and time-resolved photoluminescence (TRPL). Special concerns on the photoluminescence dynamics and correlation with structural and optical properties are discussed. This research provides useful information to optimize QW structures for various optical applications.

Experiment

The epitaxial growth of InxGa1-xN/GaN MQWs on c-plane (0001) sapphire substrates, labeled as A and B (run numbers of RN9 and RN11), were performed by MOCVD. For the growth procedures, the substrates were initially treated in a H_2 ambient at 1173 °C, followed by the growth of a low-temperature (550 °C) GaN buffer layer and a layer of nominally undoped GaN grown at high temperature (1020 °C) under low pressure. Afterwards, the temperature was lowered to grow five periods of InGaN/GaN quantum wells layer in the final step. Both samples of A and B consist of

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5-QWs with well width of 1.6 nm and barrier width of 7.9 nm. The indium concentration of sample A and B in the well are different, 24% and 21%, respectively, which also agrees with the TEM and HR-XRD simulation results.

Results and discussion

TDPL of InGaN/GaN LED samples

Fig. 1 shows the T-dependent PL spectra of the sample A and B, over a temperature range of 15 to 300 K. All spectra are normalized to have nearly the same intensity. The peak position is hard to decipher because of the Fabry-Perot effect. In order to indicate the exact InGaN PL peak position, Gaussians were used to fit the QW-related energy peak.

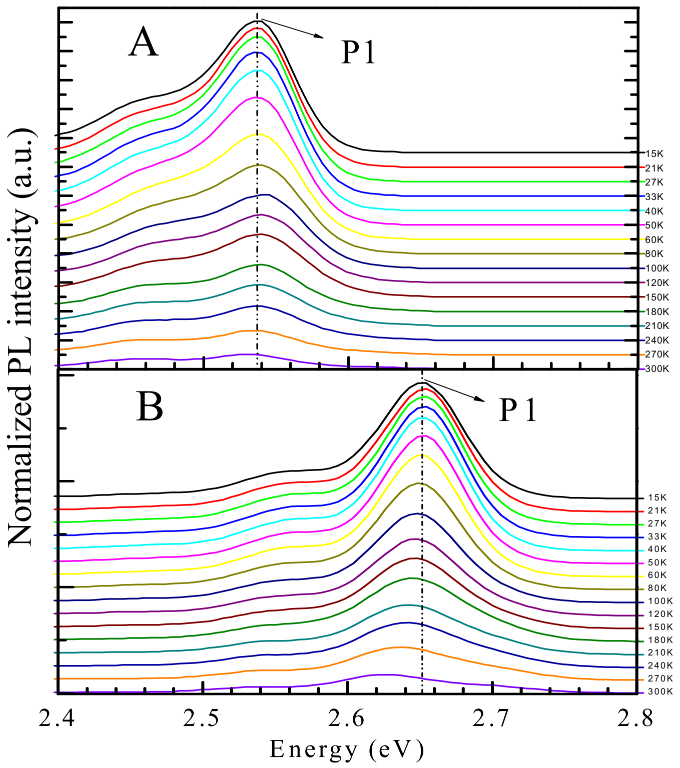

Fig. 1 Temperature-dependent PL spectra for the InGaN/GaN MQWs from 15 to 300 K.

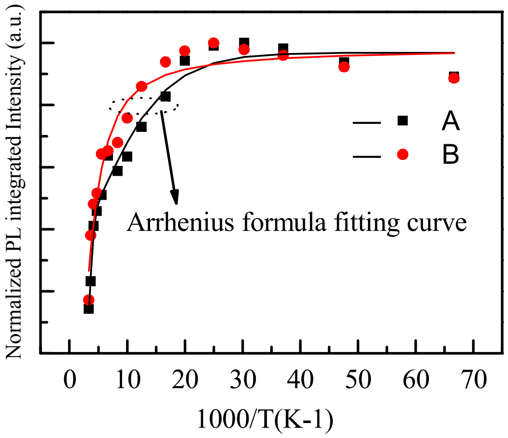

Fig. 2 Arrhenius plot of the normalized integrated PL intensity (IPL) versus temperature for sample A and B.

Tab. 1 Activation energy obtained from equation (1).

Sample names	Indium content, x	E_a (meV)	E_b (meV)
RN9	0.24	90.00	2.00
RN11	0.21	70.00	1.60

At room temperature the intensity tends to decrease with a red-shift. Such behaviors are quite different from previous results, which were usually observed in typical III–V compounds. In such a compound, the PL intensity always decays with temperature due to the increasing nonradiative recombination. Such an anomalous phenomenon can be attributed to carrier supply with thermal energy. Due to thermal energy, carriers can overcome certain potential barriers and relax into localized states formed by the InGaN clusters. With increasing temperature, carriers can obtain enough thermal energy to relax into the localized states. Therefore, the localized states gradually dominate the emission at higher temperature and the FWHM increases with temperature. The T-dependence of integrated PL intensity IPL could be expressed as [6, 7]

$$I_{PL}(T)=\frac{I_0}{1+C_1e^{(\frac{-E_a}{K_BT})}+C_2e^{(\frac{-E_b}{K_BT})}} \tag{1}$$

where I_0 is the intensity at T = 0 K, C_1 is the ratio between the exciton radiative lifetime in the QW and the exciton escape time from the QW to a nonradiative center, k_B is Boltzmann constant, E_a is the activation energy, E_b is the activation energy of the second non-radiative center, and C_2 is the efficiency of the second quenching mechanism.

Fig. 2 shows an Arrhenius plot, i.e., ln(I(T)) versus 1000/T, of the normalized integrated PL intensity for the InGaN-related PL emission over the temperature range from 15 to 300 K, which gives an anomalous temperature-dependent PL intensity of the emission peak, i.e. not monotonous decreasing with increasing temperature for sample A and B.

The good fit is obtained by using Equation (1). The obtained activation energies are listed in Table 1. The PL line shape indicates that carriers are free and thermalized for temperature above 100 K. Thus, the thermal quenching activation energy must represent either a barrier to capture at nonradiative recombination centers, or the thermal activation energy of such centers [8]. It can be seen that the data over lower temperature ranges cannot be well fitted by the Arrhenius formula. The main reason could be considered due to the existence of the localization centers and carrier thermalization effect. As the temperature increases, the carrier can be captured by other localization centers.

Photoluminescence Excitation (PLE)

Fig. 3 shows RT InGaN-MQW-related PL, and PLE spectra measured with different excitation photon energies E_{exc} (excitation energy), for the sample A and B, respectively. It is noted that the carrier generation ratio of the InGaN wells to the GaN barriers achieves the maximum when the PLE detection energy was set at the PL peak energy (curve I). When the detection energy is below the InGaN main emission peak (curve II), the contributions of the GaN regions are enhanced compared to the curve I. These facts imply that for E_{exc} (excitation energy) above $E_{g,GaN}$ (E_g of the GaN barriers), the lower-energy side of the InGaN main emission peak is governed mainly by carrier generation in the GaN barriers and subsequent carrier transfer to the InGaN wells. From the different PLE contributions for the higher and lower energy sides of $E_{p,InGaN}$, different recombination mechanisms for various excitation energies can be expected. As the PLE detection energy is monitored at (IV) 2.08 and (III) 2.278 eV for the YB emission, the contribution of the GaN barrier layer is noticeably increased. These facts indicate that the main source of the YB emission does not predominantly originate from the InGaN wells but originates from the GaN barrier layers.

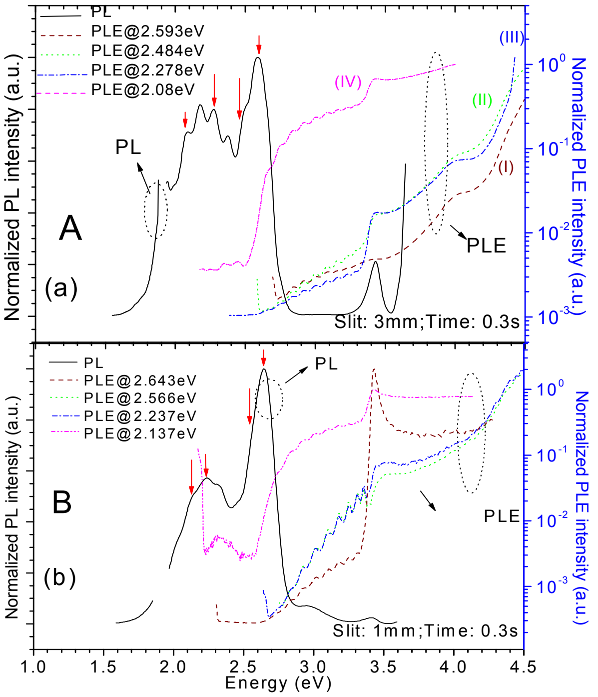

Fig. 3 PL and PLE spectra at RT of two samples. The PL measurement was excited with a xenon lamp coupled with a monochromator (325nm). PLE spectrum was taken at energies indicated by the red arrows.

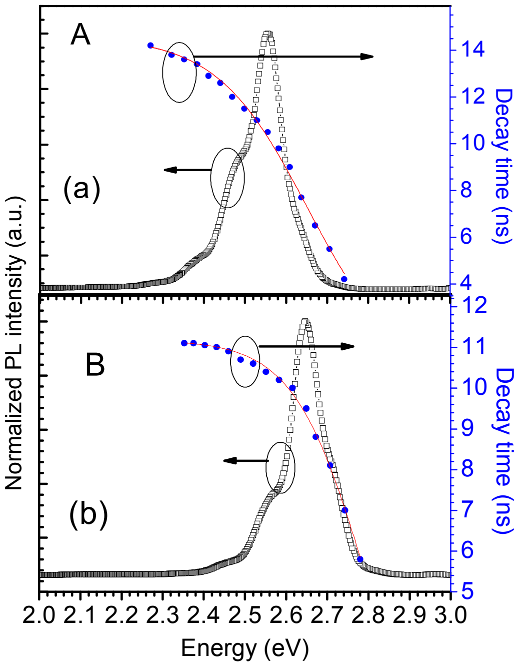

Fig. 4 Photon energy dependence of PL lifetime τ_{PL} for two samples (a) sample A between 2.27 to 2.84 eV and (b) sample B between 2.37 to 2.9 eV at room temperature. The integrated PL spectrum (open square symbols) is also shown.

In order to analyze the Stokes shift, which is defined as the difference in energy between the effective band gap and the emission peak energy, it is essential to have an accurate description of the absorption edge that includes the effects of broadening. PLE measurement was performed to get the absorption edge. Martin et al. suggested that by fitting the PLE spectra to the sigmoidal formula [9],

$$\alpha=\alpha_0/[1+\exp((E_{eff}-E)/\Delta E)] \qquad (2)$$

where α_0 is the constant, E_{eff} the effective band gap, and E the broadening parameter which indicates a distribution of absorption states, and E is the excitation energy. We obtained E_{eff} values of 2.931 and 2.884 eV, and ΔE values of 17.69 and 21.15 meV, for the samples with In compositions of 21% and 24%, respectively. This broadening of the PLE spectra with higher In indicates that the absorption states are distributed over a wider energy range due to an increase in the degree of fluctuations in dot size and/or shape [10], or due to interface imperfection as shown in Fig. 3. The Stokes shift between the PL peak energy and the ''effective band gap'' (E_{eff}-E) was 288 and 304 meV, respectively, with increasing In composition. The Stocks shift was obtained as high as 304 meV, which is consistent with the large value of activation energy associated with the strong localization of the carriers in this MQW structure.

Time-resolved Photoluminescence (TRPL)

To further clarify the emission properties of InGaN/GaN MQWs, The TRPL of sample A and B was measured at different wavelength. The photon energy dependence of τ_{PL} at RT is shown in Fig. 4. The normalized PL spectra are also shown in these figures.

It can be seen that PL has a main peak around 2.54 and 2.65 eV for sample A and B, respectively. The PL values increase with decreasing photon energy in the two samples, which was due to the energy transfer from a higher energy localized state to lower one. This is characteristic of the localized system, where the decay of excitons consists of both radiative recombination and the transfer process to tail states. The depth of localization can be evaluated by assuming the exponential distribution of the density of tail states and by fitting the photon energy dependence of the τ_{PL} values using the following equation [11]:

$$\alpha=\tau_{rad}/[1+\exp((E-E_{me})/E_0)] \qquad (3)$$

where τ_{rad}, E_{me}, and E_0 are the radiative lifetime, the energy similar to the mobility edge, and the depth of localization, respectively. The radiative lifetime, the energy similar to the mobility edge, and the depth of localization obtained from Equation (3) are obtain. By fitting, τ_{rad} = 14.60 ns, E_{me} = 2.65 eV, and E_0 = 109.98 meV were obtained for sample A, while τ_{rad} = 11.80 ns, E_{me} = 2.79 eV, and E_0 = 85.88 meV were for sample B. The lifetime at the low-energy side of the InGaN peaks is longer for higher indium composition, as expected from the larger Stokes shift [12].

At room temperature, the samples have different lifetimes because of thermally activated nonradiative recombination. In this case, the majority of excitons in the InGaN QWs on sample B is delocalized from the shallow potential minima and recombines radiatively at around RT. On the other hand, InGaN QWs on sample A have a deeper localization depth of 109.98 meV. These results indicate the suppression of nonradiative recombination for higher indium composition samples. The possible mechanism for these phenomena can be argued as follows. First, the effect of localization keeping carriers away from nonradiative pathways can be enhanced with increasing indium and thereby increasing the Stokes shift. Second, the incorporation of more indium into the InGaN well layer can reduce the density of nonradiative recombination centers

Summary

In conclusion, we have studied the effect of different indium compositions on the structural and optical properties for two InGaN MQW samples. As the temperature increases, carriers transfer between different luminescent centers. From the calculation of the activation energy, abnormally high activation energy was obtained, which shows that the excitons are not in the free states. The large Stokes shift between PL and absorption peak can be attributed to the variation in indium composition or the QCSE. PLE spectra of different detection energy reveal that the lower-energy side of the InGaN main emission peak is governed mainly by carrier generation in the GaN barriers and the subsequent carrier transfer to the InGaN wells, which leads to a longer radiative lifetime, a bigger activation energy, a deeper localization potential, and a larger Stokes shift for the low-energy side of the InGaN peaks with higher indium composition. It can therefore be concluded that the use of adjusting the indium composition is good method to improve the optical efficiency of InGaN-based LEDs.

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