Influence of growth temperature on emission efficiency of InGaN/GaN multiple quantum wells

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ABSTRACT

A comparative study, using time-resolved and CW photoluminescence spectroscopy, of MOVPE grown InGaN/GaN multiple quantum wells deposited on HVPE GaN/Sapphire at different growth temperatures was undertaken. It was found that the PL linewidth increased and the peak emission energy decreased as the growth temperature was reduced. Moreover, the sample grown at an intermediate growth temperature exhibited total integrated luminescence intensity much greater than the samples grown at higher or lower growth temperatures. A phenomenological carrier recombination dynamics model based on the competition of quantum well-like radiative recombination, spatially localized radiative recombination in potential minima and non-radiative recombination through defects is presented to provide an explanation of the observed emission dynamics and efficiency. In this model, the emission efficiency is determined by the relative area of defects and the number density of localized states in the potential minima, both of which are influenced by the growth temperature. Furthermore, the photon energy dependent lifetimes are well fitted with this model by assuming a Gaussian shape localized states distribution. The localized potential minima are consistent with nanoscale indium rich regions due to indium aggregation.

INTRODUCTION

It is striking to realize that although InGaN has emerged in the last few years as one of the most important materials for short-wavelength optoelectronics the mechanisms of radiative recombination in InGaN QWs is still subject to debate in the literature. It has been recognized that under typical growth conditions there is a positive enthalpy for indium mixing in GaN. Moreover, electromicroscopy and cathodoluminescence of InGaN have demonstrated the existence of nanometer and micron scale regions of high indium concentration. It has been hypothesized that the ‘quantum dot’ like nanoscale regions of high indium concentration prevent the carrier capture from the non-radiative defects and improve efficiency of light-emitting diodes. However, the effects of growth conditions on indium phase segregation, which affect the carrier recombination dynamics and device emission efficiency, although acknowledged, remain unclear due to the complex nature of growth processes in III-N materials. Clearly, one of the most important parameters for growth of high quality InGaN thin films is growth temperature.

In this article, we present femtosecond time-resolved and CW photoluminescence spectroscopy to compare emission from MOVPE grown InGaN-based MQWs deposited on HVPE GaN/Sapphire at three different growth temperatures but with otherwise identical conditions. Moreover, a phenomenological carrier recombination dynamics model based on the competition of quantum well-like radiative recombination, localized radiative recombination in potential minima and non-radiative recombination through defects is presented to provide an explanation of the observed emission dynamics and efficiency.
SAMPLES AND EXPERIMENTS

A set of In\(_{0.1}Ga_{0.9}\)N/GaN MQWs were deposited at different growth temperatures by MOVPE on HVPE GaN/Sapphire substrates. The indium composition was nominally 10% in all the MQW samples. The MQW structure consists of six layers of InGaN 70 Å thick wells alternating with seven layers of GaN barriers each 90 Å thick. Detailed temperature and excitation power dependent time-resolved and time-integrated photoluminescence measurements were performed on this sequence of samples.

![Figure 1. Time-integrated PL spectra for MQW A, B and C taken at 18 K, as shown in curve A, curve B and curve C. The PL intensity for MQW C is multiplied by a factor of 7.2, and for MQW A is multiplied by a factor of 1.17.](image)

Typical time–integrated PL spectra of the three different InGaN/GaN MQWs at 18 K are shown in Figure 1. Clearly, the optical properties of the three InGaN/GaN MQW structures are very dependent on the growth temperature. This is reflected in the fact that for A (high growth temperature), B (intermediate growth temperature), and C (low growth temperature), the PL peak energy varies from 3.221 eV, 3.094 eV to 2.948 eV, and the full width at half-maximum (FWHM) of emission increases from 49 meV, to 64 meV and finally to 148 meV respectively. These results are consistent with the increasing indium incorporation efficiency and an increasing magnitude of nanoscale fluctuations in the localization with decreasing growth temperature. More importantly, sample B grown at an intermediate growth temperature, exhibited a total integrated luminescence intensity, which is 1.54 times and 2.98 times of the samples grown at high or low growth temperature respectively.

THEORETICAL MODELLING

It has been reported that the emission in InGaN/GaN MQWs is from localized excitonic radiative recombination in the indium rich potential minima due to indium fluctuations, and the presence of quantum dot like indium rich regions, although it reduces crystalline quality, provides an efficient radiative trap for carriers.\(^7\)\(^-\)\(^9\) In order to understand the growth temperature dependent emission efficiency of InGaN/GaN quantum well structures, a model based on the above hypothesis is proposed. Figure 2 shows an energy level diagram with representative levels
for the quantum well emission, for a spatially localized emission in the potential minima and finally a level that represents the defects within the system.

![Energy Diagram](image)

Figure 2. Carrier recombination dynamics model in InGaN quantum well. Efficiency is affected by diffusion and drift to both defect regions and localized potential minima.

The localized radiative recombination in the indium rich regions is influenced by the motion of the photon-generated carriers among the different localized energy states, and by the competition between the defects and localized states. To understand these processes in more detail, the time evolution of the photo-generated carrier density in the different regions can be described by the following rate equation.

\[
\frac{dN_{QW}}{dt} = -\frac{N_{QW}}{\tau_{QW-rad}} - \frac{N_{QW}}{\tau_{QW-nr}} - \frac{N_{QW}}{\tau_{Drift-LS}} - \frac{N_{QW}}{\tau_{Drift-Defect}} + \frac{I(t)}{h\nu}
\]

\[
\frac{dN_{LS}(E)}{dt} = \frac{N_{LS}(E)}{\tau_{LS-rad}} + \frac{N_{QW}}{\tau_{QW-rad}} - \frac{\sum Tr(E \rightarrow E') N_{LS}(E')}{\tau_{LS-nr}} - \frac{\sum Tr(E \rightarrow Defect) \times N_{LS}(E)}{\tau_{LS-nr}}
\]

\[
\frac{dN_{Defect}}{dt} = -\frac{N_{Defect}}{\tau_{Defect-nr}} - \frac{N_{QW}}{\tau_{Drift-Defect}} + \sum_{E} Tr(E \rightarrow Defect) \times N_{LS}(E)
\]

where \(N_{QW}, N_{LS}(E)\) and \(N_{Defect}\) represent the carrier density in the quantum well, in the spatially localized potential minima with different localization energy \(E\) and in the defects respectively. The recombination dynamics in the quantum well and potential minima is characterized by the radiative lifetimes \(\tau_{QW-rad}, \tau_{LS-rad}\) and the non-radiative lifetimes \(\tau_{QW-nr}, \tau_{LS-nr}\) respectively.

Gourdon and Lavallard have proposed that the transfer probability of excitons created by optical excitation in a localized state in CdS_{1-x}Se_{x} alloys was proportional to the number of states available at the lower energy. In a similar manner, we define \(Tr(E \rightarrow E')\) and \(Tr(E \rightarrow Defect)\) as the transfer rates from the localized energy \(E\) state to the localized energy \(E'\) state and from the localized state to the surrounding defect regions. At low temperatures, the transfer rate \(\sum_{E < E'} Tr(E \rightarrow E')\) is considered to be proportional to the total number density available at the
lower localized energy states \[ \int_{E}^{+\infty} g(E') dE' \] (E is the localization energy), where the density of states \( g(E') \) is chosen to be Gaussian shape. Likewise, the transfer rate \( \text{Tr}(E \rightarrow \text{Defect}) \) is proportional to \( n_{\text{Defect}} \times v \times \sigma_{\text{Defect}} \), where \( n_{\text{Defect}} \) is the defect density, \( v \) is the thermal velocity, and \( \sigma_{\text{Defect}} \) is the cross section of the defect. The relaxation rates \( 1/\tau_{\text{Drift-Defect}} \) and \( 1/\tau_{\text{Drift-LS}} \) would also be proportional to the \( n_{\text{Defect}} \times v \times \sigma_{\text{Defect}} \) and \( n_{\text{LS}} \times v \times \sigma_{\text{LS}} \) respectively. Using these approximations, the photon energy dependent lifetime in the localized potential minima and the total emission efficiency from the localized potential minima can be given by

\[
\frac{1}{\tau} = \frac{1}{\tau_r} + \text{Tr}(E \rightarrow \text{Defect}) + \sum_{E < E'} \text{Tr}(E \rightarrow E') = \frac{1}{\tau_r} + \alpha \times \int_{E}^{+\infty} g(E') dE' + \beta \times n_{\text{Defect}} \times v \times \sigma_{\text{Defect}} \tag{2}
\]

\[
\eta = \frac{1}{1 + \frac{n_{\text{Defect}} \times \sigma_{\text{Defect}}}{n_{\text{LS}} \times \sigma_{\text{LS}}} \times \frac{1}{1 + \frac{\beta \times n_{\text{Defect}} \times v \times \sigma_{\text{Defect}} \times \tau_{\text{LS-rad}}}{1}} \tag{3}
\]

where \( \alpha \) and \( \beta \) are fitting parameters. The above equation for efficiency indicates that reducing defect density and increasing the localized states are all favorable to improve radiative efficiency. Thus an optimal growth condition is suggested to get both optimal localization and high crystalline quality when fabricating highly efficient InGaN/GaN quantum well emitters.

**RESULTS AND DISCUSSION**

For simplicity and clarity, Eq. 3 can also be expressed as \( \eta = \eta_1 \times \eta_2 \), where

\[
\eta_1 = \frac{1}{1 + \frac{n_{\text{Defect}} \times \sigma_{\text{Defect}}}{n_{\text{LS}} \times \sigma_{\text{LS}}} \times \frac{1}{1 + \beta \times n_{\text{Defect}} \times v \times \sigma_{\text{Defect}} \times \tau_{\text{LS-rad}}}}, \quad \text{and} \quad \eta_2 = 1/\left(1 + \beta \times n_{\text{Defect}} \times v \times \sigma_{\text{Defect}} \times \tau_{\text{LS-rad}}\right). \]

We use the emission intensity (integrated over the entire emission wavelength) near zero delay after a femtosecond laser pulse excitation in the time-resolved photoluminescence spectra to estimate the relative area ratio between the efficient localized radiative recombination centers and the defects, provided carriers generated in the quantum well are distributed into the localized potential minima and defects evenly. Hence, the ratio of \( \eta_1 \) determined by the \( (n_{\text{Defect}} \times \sigma_{\text{Defect}})/(n_{\text{LS}} \times \sigma_{\text{LS}}) \) in these three samples is given by \( \eta_1(A):\eta_1(B):\eta_1(C) = 1:3.27:1.41 \). If \( \sigma_{\text{Defect}} \) and \( \sigma_{\text{LS}} \) are assumed to keep constant, this indicates the density ratio \( n_{\text{Defect}}/n_{\text{LS}} \) will increase as the growth temperature is reduced or raised from an intermediate growth temperature, and in what follows there exists an optimal growth temperature where the \( n_{\text{Defect}}/n_{\text{LS}} \) can reach a minimum value.

Eq. 2 indicates that the carrier lifetime at different localized energy states is determined by the radiative recombination lifetime, the defect density and the density of states distribution in the spatially localized region. If the photo-generated carriers in the quantum well will diffuse to the potential minima based on the localized density of state distribution immediately after pulse excitation, the time-resolved emission spectra at short times reflects the true distribution of the density of states over the whole emission wavelength. As shown in Figure 3(b), Gaussian like distributions are obtained by integrating the time-resolved PL intensity from 0 ps to 200 ps. The density of states distribution represented by the dotted curve in Figure 3(b) is used to fit the photon energy dependent lifetime. The photon energy dependent lifetimes for these samples are...
fitted by using Eq. 2, as shown in Figure 3(a). The fitting results estimated the transfer lifetimes from localized states to defects as 12 ns, 2.069 ns and 0.845 ns for sample A, B and C respectively. The localized radiative recombination lifetime is estimated to be the same, 1.5 ns. The resulting efficiency $\eta_2$ can be obtained as $\eta_2(A):\eta_2(B):\eta_2(C)=1:1.6:0.41$. Therefore, the final total efficiency is calculated by $\eta = \eta_1 \times \eta_2$ and given as $\eta(A):\eta(B):\eta(C)=0.469:1.0:0.267$. It is clear that these results are very close to the experimental integrated luminescence efficiency ratios of 0.651:1:0.335.

![Figure 3](image_url)

Figure 3. (a) Photon energy dependent lifetimes of sample A, B and C at 15 K. (b) Distribution of density of states and integrated density of states for sample A, B and C.

It has been reported that the mean diameter of self-assembled InGaN quantum dots is reduced and the indium content increased by decreasing the growth temperature. This is because the low growth temperature suppresses the migration of In and Ga atoms. On the other hand, the standard deviation of quantum dot size decreases monotonically as the growth temperature is raised. Moreover, it is well known the growth of InGaN alloys at high temperature results in high crystalline quality but low indium concentration because of the high volatility of N over InN.1 Applying these results into the spatially localized indium rich potential minima in our case where the localization energy is determined by the indium concentration and potential minima size, we achieve excellent agreement with the peak energy red shift and the increase of FWHM from sample A to sample C as shown in Fig. 1.

Eq. 1 suggests the emission from the quantum well will be replaced with the emission from the spatially localized potential minima, as the density of indium rich potential minima increases when lowering the growth temperature. It will improve the overall device emission efficiency. However, if the growth temperature is too low, the degrading crystalline quality with high defect density will finally decrease the efficiency observed in sample C. Therefore, an optimum growth temperature is suggested to reach the maximum efficiency and it can be
estimated from Eq. 3. In order to determine this optimum growth temperature, more investigations of the effects of the growth conditions are needed to better understand the competition between the non-radiative recombination through defects and the localized radiative recombination in potential minima. In addition, a better understanding of the formation of indium rich region and defect propagation versus growth temperature is needed.

CONCLUSION

In this paper, we have presented experimental studies of a set of InGaN/GaN MQWs structures and provided a description of spatially localized potential minima due to phase segregation in InGaN quantum well as well as the resulting carrier dynamics. PL spectra reveal red-shifts and decreasing linewidth with reducing growth temperature. The sample grown at an intermediate growth temperature exhibited the highest integrated luminescence efficiency. These results are consistent with decreasing indium incorporation efficiency and decreasing indium fluctuation with increasing growth temperature. A carrier recombination dynamics model is presented and is able to account for the variety of luminescence results of these samples. This interpretation is supported by the observation that the luminescence characteristics are determined by the competition among quantum well radiative recombination, spatially localized radiative recombination, and non-radiative recombination in the defects. The photon energy dependent lifetimes are well fitted with this model by assuming a Gaussian localized states distribution. According to this model, the existence of an optimum growth temperature is suggested to get the maximum device efficiency and it is also verified by the experiment results.

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