# Ultrafast Spectroscopy of InGaN Quantum Wells for the Development of Efficient Emitters

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**Abstract**

**Ultrafast time-resolved and CW photoluminescence (PL) measurements are used to study the recombination mechanism in MOVPE grown InGaN/GaN multiple quantum wells (MQWs) at different growth temperatures. It was found that the emission efficiency is determined not only by the defect density but also by the number density of localized states in the potential minima. A phenomenological model is proposed to evaluate the competition between these two factors and to estimate the emission efficiency by using the time-resolved PL experiment results. The localized potential minima are consistent with nanoscale indium rich regions due to indium aggregation.**

## INTRODUCTION

In the past few years, InGaN-based light emitting diodes from ultraviolet to amber and blue/ultraviolet laser diodes have been fabricated successfully, in spite of high dislocation densities. Many authors hypothesized that the 'quantum dot' or 'quantum disk' like nanoscale regions of high indium concentration in the active layers prevent the carrier capture from the non-radiative defects and improve efficiency of light-emitting diodes.[1,2] Moreover, electromicroscopy and cathodoluminescence of InGaN have demonstrated the existence of nanometer to micron scale regions of high indium concentration.[3-5] However, the effects of growth conditions on indium phase segregation, which affect the carrier recombination dynamics and device emission efficiency, although acknowledged,[6] remain unclear due to the complex nature of growth processes in III-N materials.

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 by MOVPE on HVPE GaN/Sapphire substrates with different growth temperatures. 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 PL measurements were performed on this sequence of samples.
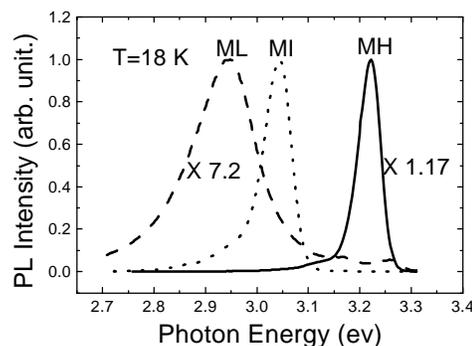


Figure 1. Time-integrated PL spectra for MH (solid line), MI (dotted line), and ML (dashed line) grown at different growth temperatures taken at 18 K. The PL intensity for ML is multiplied by a factor of 7.2, and for MH is multiplied by a factor of 1.17.

Typical time–integrated PL spectra of MH (high growth temperature), MI (intermediate growth temperature), and ML (low growth temperature) 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 MH, MI and ML, 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, MI grown at an intermediate growth temperature, exhibited a

total integrated luminescence intensity, which is 1.537 times and 2.983 times of the samples grown at high or low growth temperature respectively.

## MODELING

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.
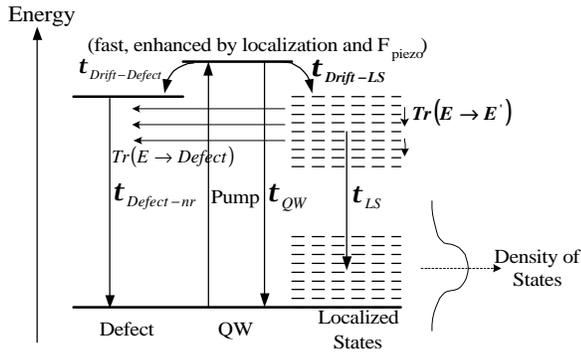


Figure 2. Carrier recombination dynamics model in InGaN quantum well. Efficiency is affected by diffusion and drift to both defect regions and localized potential minima.

First, the photo-generated carriers created in the quantum well will drift to the localized states or the non-radiative defects at a very fast time scale. The drift rates, $1/\hat{o}_{\text{Drift-Defect}}$ and $1/\hat{o}_{\text{Drift-LS}}$, are determined by the defect density $n_{\text{Defect}}$ and the number density of localized states $n_{\text{LS}}$ respectively. Then, the carriers in the localized state can emit photons through radiative recombination, relax to the lower energy localized states, or transfer to the defect regions. The transfer rates from a localized energy state to another lower localized energy state is considered to be proportional to the total number density available at this lower localized energy state. Likewise, the transfer rate from the localized state to the surrounding defect regions is proportional to defect density. Using these approximations, the total emission efficiency from the localized potential minima can be given by

$$h = h_1 \times h_2 = \frac{1}{1 + a \times \dfrac{n_{Defect}}{n_{LS}}} \times \frac{1/t_{LS}}{1/t_{LS} + b \times n_{Defect}} \quad (1)$$

where $á$ and $â$ are fitting parameters, $\hat{o}_{\text{LS}}$ is the radiative lifetime in the localized state. The first term, $\varsigma_1$, indicates the ratio of the photo-generated carriers in quantum well drifting to the localized states. The second term, $\varsigma_2$, is the ratio of the carriers recombining radiatively in the localized states. The above equation suggests, as expected, reducing defect density increases the device efficiency. In addition, this equation suggests that increasing the localized states is also favorable to improving device efficiency. Thus an optimal growth condition is one that would get both optimal localization and high crystalline quality when fabricating high efficient InGaN/GaN quantum well emitters.

## DISCUSSION

As shown in Figure 3, the emission intensity (integrated over the entire emission wavelength) near zero delay after a femtosecond laser pulse excitation in the time-resolved PL spectra can be used to estimate $\varsigma_1$ (since near zero time we are seeing emission before transport processes can occur). This results in the ratio of efficiencies given as $\varsigma_1(MH):\varsigma_1(MI):\varsigma_1(ML)=1:3.27:1.41$. This indicates that the density ratio between defects and localized states will increase as the growth temperature is reduced or raised from an intermediate optimum growth temperature.
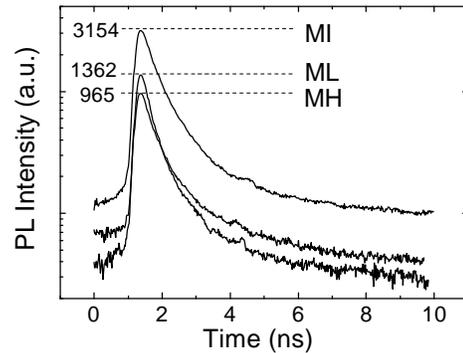


Figure 3. Time resolved emission intensity integrated in the whole emission wavelength for MH, MI and ML at 15 K.

If the photo-generated carriers in the quantum well will diffuse to the potential minima based on the density of localized 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. Figure 4(a) shows the Gaussian like distributions obtained by integrating the time-resolved PL intensity from 0 ps to 200 ps. The photon energy dependent lifetimes for these samples are fitted by the model, as shown in Figure 4(b). The fitting results estimated the transfer lifetimes from localized states to defects as 12 ns, 2.069 ns and 0.845 ns for sample MH, MI and ML respectively. The localized radiative recombination lifetime is estimated to be the same, 1.5 ns. The resulting efficiency

$\varsigma_2$ can be obtained as $\varsigma_2(MH):\varsigma_2(MI):\varsigma_2(ML)=1:1.6:0.41$. Therefore, the final total efficiency is calculated by $\boldsymbol{h}=\boldsymbol{h}_1\times\boldsymbol{h}_2$ and given as $\varsigma(MH):\varsigma(MI):\varsigma(ML)=0.469:1:0.267$. It is clear that these results are very close to the experimental integrated luminescence efficiency ratios of 0.651:1:0.335.
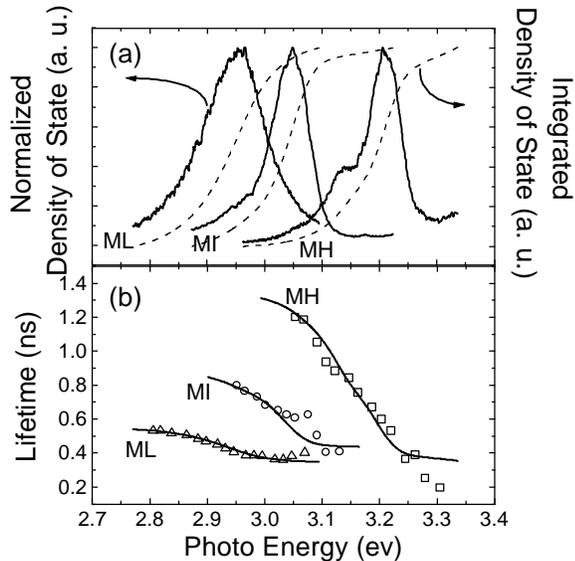


Figure 4. (a) Distribution of density of states (solid curve) and integrated density of states (dashed curve) for MH, MI and ML at 15 K; (b) Photon energy dependent lifetimes of MH, MI and ML at 15 K. Solid line is the fit from the model.

In this model, we discuss nanoscale regions of lower potentials that enhance radiative efficiency. Of course, the next extension of this would be the growing of self-assembled InGaN quantum dots. It has been reported that the mean diameter of quantum dots is reduced and the indium content increases 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.[10,11] Moreover, it is well known that the growth of InGaN alloys at high temperature results in high crystalline quality but with low indium concentration because of the high volatility of N over InN.[6] 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 MH to ML as shown in Figure 1.

When lowering the growth temperature, 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. This 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 ML. Therefore, an optimum growth temperature exists to reach the maximum efficiency and it can be estimated from Eq. 1 once the dependence of the defects and localized states on growth temperature are completely determined. In order to determine this optimum growth conditions, more investigations of the effects of the growth processes 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.

CONCLUSIONS

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 redshifts and decreasing linewidth with reducing growth temperature. The sample grown at an intermediate growth temperature exhibited the highest integrated luminescence efficiency. 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. It is able to provide useful guidance for the development of growth for high efficient III-Nitrides emitter.

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ACRONYMS

PL: Photoluminescence

MQW: Multiple Quantum Well

FWHM: Full Width at Half Maximum