Optical Characterization of Carrier Localization, Carrier Transportation and Carrier Recombination in Blue-Emitting InGaN/GaN MQWs

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Carrier localization, transportation and recombination in blue-emitting InGaN/GaN multiple quantum wells were analyzed using temperature-dependent photoluminescence spectroscopy, confocal laser scanning microscopy and time-resolved photoluminescence (TRPL). The temperature-dependent shift of PL intensity was fitted with Arrhenius equation and explained using two non-radiative channels, which are related with thermal activation of carriers from different confining potentials. The S-shaped shift of PL peak energy and inverse-S-shaped shift of PL full width at half maximum were explained with carrier localization and carrier transportation. The TRPL spectra taken at several different places from bright region to dark region in the confocal microscopic image showed that the fast decay life-time $\tau_1$ increases with decreasing PL intensity, indicating a higher carrier transportation rate at bright region, while the slow decay life-time $\tau_2$ decreases with decreasing PL intensity, indicating a higher probability of non-radiative recombination at dark region.

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Experimental

Two InGaN/GaN MQWs samples were grown on c-plane sapphire substrate in a Veeco K465i GaN metal-organic chemical vapor deposition (MOCVD) reactor. Triethylgallium (TEGa) and trimethylindium (TMIn) were used as group III sources. Ammonia (NH$_3$) was used as group V source. Nitrogen was used as carrier gas. A 30-nm-thick GaN nucleation layer was grown first on the substrate, followed by a 0.5-μm-thick GaN buffer layer and a 2-μm-thick Si-doped n-type GaN layer. This preliminary structure serves as a GaN template for further growth. Analysis of the GaN template is as follow. The X-ray diffraction (XRD) $\omega$(002) and $\omega$(102) rocking curves scan indicated a TD density of approximately 4.4×10$^6$ cm$^{-2}$. The atomic force microscopy (AFM) surface morphology scan of the GaN template in a 5×5 μm$^2$ area exhibited a root mean square (RMS) roughness of 0.4 nm. Four-period MQWs were then grown on the GaN template, consisting of 2.7-nm-thick InGaN quantum well layers and 12.5-nm-thick GaN barrier layers, as was confirmed by XRD. Detailed growth conditions are described elsewhere.$^5$ The growth temperature was also tuned to achieve a target PL peak wavelength of $\sim$449 nm, corresponding to an indium composition of $\sim$14%.

Figure 1 shows the system diagram of low-temperature PL system. A Verdi-G10-semiconductor-laser pumped laser system, generating 400 nm CW laser beam, was used as excitation source. The laser beam, after transmitting through an optical fiber, was focused into a 2-mm-diameter spot on the sample. PL light from sample was detected by monochromator with Si detector. The sample was mounted on a cold finger in a Cryo-head, which was connected with a vacuum pump, a Helium compressor and a temperature controller, so that temperature between 10 K and 300 K can be reached. A computer was connected to the system, and a Labview program was written to control the temperature controller as well as to acquire data from monochromator and Si detector, so that the PL spectrum can be taken at any temperature between 10 K and 300 K.

For nanometer scale imaging and TRPL measurement, a CLSM system, integrated with TRPL measurement system, was used. A Pico-Quant laser diode, generating 402 nm pulsed laser beam with 10 MHz frequency and 300 ps pulse width (0.3% duty cycle), was employed as excitation source. The PL emission was detected by a time-correlated single photon counting (TCSPC) avalanche photodiode (APD), which was preceded by a 445/40 nm band pass filter so that only near-bandedge emission contributes to the imaging. The microscope had a lateral spatial resolution of 200 nm and vertical spatial resolution of 100 nm. Average laser power density on the sample was 5.1 W/cm$^2$, indicating an average carrier injection density of 1×10$^{17}$ cm$^{-3}$. This excitation condition was low enough to avoid sample heating. Details of the CLSM and TRPL system are described elsewhere.$^5$

Results and Discussion

Temperature-dependent PL.— Figure 2 shows the PL spectra of sample #1 measured at different temperatures. The low energy shoulder corresponds to longitudinal optical phonon replica of the main recombination peak, which is proved by the fact that it disappears at increased temperature. The high energy shoulder, on the other hand, does not appear until temperature is higher than 100 K. It corresponds to carrier recombination at weakly localized state, since carriers cannot be thermal activated to higher energy level unless the temperature is high enough.

In Figure 3 the spectrum-integrated PL intensity is plotted as a function of the temperature. It is seen that the spectrum-integrated PL intensity decreases with increasing temperature, indicating that more carriers recombine non-radiatively at higher temperature. Curve fitting done on the data in Figure 3, based on first-order standard Arrhenius equation which assumed only one non-radiative channel, did not result in good coherent to the experiment. However, fitting

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Based on second-order Arrhenius equation

\[ I(T) = \frac{I_0}{1 + A \exp(-E_A/k_B T) + B \exp(-E_B/k_B T)}, \]

which assumes two non-radiative channels with rate constant \( A \) and \( B \) and activation energy \( E_A \) and \( E_B \), result in good coherent with experiment data, by setting \( A = 15.6, B = 1.3, E_A = 44.4 \text{ meV} \) and \( E_B = 11.3 \text{ meV} \), as shown in Figure 3. The contributions of each non-radiative channel are plotted separately in Figure 3. It is seen that at high temperature the non-radiative recombination is dominated by the first channel (with higher activation energy \( E_A \)), while at relatively low temperature the second non-radiative channel (with lower activation energy \( E_B \)) dominates the non-radiative recombination process. The first channel corresponds to thermal activation of carriers out of the confining potential (strongly localized state) in QW layer. The second non-radiative channel, on the other hand, corresponds to thermal activation of carriers out of weakly localized state.

PL peak energy shifts when temperature increases from 10 K to 300 K, as shown in Figure 4a. However, different from what is predicted by Varshni’s empirical expression:

\[ E_p(T) = E_p(0) - \frac{\alpha T^2}{(T + \beta)} \]

which states that as temperature increases semiconductor’s bandgap energy shrinks, the PL peak energy in Figure 4a shows an S-shaped red-blue-redshift curve, which can be divided into three parts:

1. Below 70 K, PL peak energy decreases with increasing temperature;
2. From 70 K to 160 K, PL peak energy increases with increasing temperature; and
3. Above 160 K, PL peak energy again decreases with increasing temperature. Moreover, the full width at half maximum (FWHM) of the PL spectra shows an inverse-S-shaped shift with increasing temperature, as shown in Figure 4b.

The reason for this S-shaped shift of PL peak energy and inverse-S-shaped shift of PL FWHM can be explained using carrier localization at confining potential and carrier dynamics, as shown in Figure 5.

1. When temperature is below 70 K, the radiative recombination dominates the recombination process, so the radiative recombination life-time \( \tau_r \) is shorter than the non-radiative recombination life-time \( \tau_{nr} \). The radiative recombination life-time can be expressed as:

\[ \tau_r^2D \propto \frac{1}{E_B^2} \cdot \frac{M}{\mu} \cdot \frac{\Delta(T)}{-\exp(-\Delta(T)/k_B T)} \]

where \( E_B^2 = 4\hbar^2/\mu \) is the quasi 2D exciton binding energy, \( a_B^2 = (m_d/\mu) a_H^2 \) is the 2D exciton Bohr radius, \( \Delta(T) \) is the exciton linewidth at finite temperature, and all other parameters have their usual meaning. Basically, this radiative recombination life-time is temperature dependent for the \( \Delta(T) \) and \( k_B T \) terms in Equation 3. For temperature below 70 K, \( \Delta(T) \) is much smaller than \( k_B T \). So Equation 3 can be simplified into

\[ \tau_r^2D \propto \frac{1}{E_B^2} \cdot \frac{M}{\mu} \cdot k_B T. \]

As a result, the radiative recombination life-time is proportional to temperature. Since the carrier life-time \( \tau \) is calculated by

\[ 1/\tau = 1/\tau_r + 1/\tau_{nr}, \]

it is predominantly affected by the smaller life-time. Consequently, as the temperature increases from 10 K to 70 K, the radiative recombination life-time increases, leading to larger carrier life-time.

As their life-time increases, carriers can diffuse down deeper into the confining potential before recombination, as shown in Figure 5 as step (A) and (B). This reduces the recombination energy, and as a result leads to a redshift in the peak energy with increasing temperature. Moreover, as the life-time increases, carriers can diffuse to more energy states, which increases the number of available energy states during recombination, and increases the PL FWHM.

2. When temperature is between 70 K and 160 K, non-radiative process becomes dominant during carrier recombination, and

Figure 1. Schematic diagram of low-temperature PL system.

Figure 2. PL spectra at different temperatures.

Figure 3. Spectrum-integrated PL intensity vs. temperature, and its curve fitting based on second-order Arrhenius equation. The two dash line shows separately the contribution from two nonradiative channels.
non-radiative recombination life-time is shorter than radiative recombination life-time, so carrier life-time is dominated by nonradiative recombination life-time. Since nonradiative recombination life-time is related with temperature by

\[ \tau_{nr} = \frac{1}{sN_T \sqrt{8k_BT/\pi m^*}}, \]  

where \( s \) is carrier capture cross section, \( N_T \) is the density of NRRCs, it decreases when temperature increases. Detailed calculation shows that when temperature increased from 70 K to 160 K, the nonradiative recombination life-time decreases by about 18% of the \( \tau_{nr} \) at 10 K. Consequently, carrier life-time decreases with increasing temperature at 70 K to 160 K.

With increasing temperature, carriers have shorter life-time so that they cannot diffuse to the potential minimum before recombination, as shown in Figure 5 as step (C). This leads to a blueshift in the peak energy as temperature increases. On one hand, the smaller life-time reduces the number of energy states that the carriers can diffuse to; on the other hand, the higher temperature provides more thermal energy and increases the available energy states that the carriers can reach. These two factors balance each other, leading to the fact that the PL FWHM stays almost unchanged.

As a result, the PL peak blueshift behavior caused by shorter carrier life-time becomes insignificant. On the other hand, the temperature-induced bandgap shrinkage effect, introduced by Equation 1, becomes relatively significant in this temperature range. Calculation shows that the bandgap energy shrinks by 46.1 meV when temperature increases from 160 K to 300 K, which is twice as large as the 18.3 meV bandgap shrinkage when temperature increases from 70 K to 160 K. This shrinkage in bandgap energy leads to the redshift in PL peak energy.

At higher temperature, more thermal energy are provided to the carriers, so that they can hop between energy states within a much larger energy range, which increases the available number of energy states during recombination. As a result, the PL FWHM increases.

**CLSM imaging and TRPL spectroscopy.**— Figure 6 shows the CLSM image of the InGaN/GaN MQW sample. As analyzed before, the bright regions are on the order of micrometers in size, and have smaller bandgap energy than the dark regions, due to local higher indium concentration or larger quantum well thickness. So carriers are localized in small bandgap energy bright regions.

TRPL spectra were taken at eight points, along the line shown in Figure 6, from bright region to dark region nearby. The TRPL spectra were fit with double exponential function and the resulting life-time data are listed in Table I. Table I also lists the PL peak energy at each point. Almost all places are characterized by a fast decay with small life-time and a slow decay with large life-time. However, the place with the highest PL intensity (point A) is characterized by only slow

![Figure 6](image-url)

Figure 6. CLSM image showing near-bandedge emission. Color bar shows the near-bandedge emission intensity of pixels in the image. TRPL spectra were taken from bright region (point A) to dark region (point H).
Table I. Pixel PL peak energy, intensity and life-time data.

| Point where TRPL was taken | PL peak energy at the point (eV) | PL intensity at the point (a.u.) | Lifetime $\tau_1$ (ns) | Lifetime $\tau_2$ (ns) |
|---------------------------|---------------------------------|---------------------------------|------------------------|------------------------|
| A                         | 2.7563                          | 31                              | N/A                    | 28.08                  |
| B                         | 2.7617                          | 27                              | 3.36                   | 26.97                  |
| C                         | 2.7649                          | 26                              | 3.43                   | 26.20                  |
| D                         | 2.7677                          | 23                              | 3.47                   | 25.85                  |
| E                         | 2.7744                          | 21                              | 3.82                   | 25.28                  |
| F                         | 2.782                           | 18                              | 4.01                   | 24.27                  |
| G                         | 2.7864                          | 17                              | 4.12                   | 23.05                  |
| H                         | 2.7932                          | 15                              | 4.14                   | 22.86                  |

Decay, with no observation of fast decay. The fast decay with small life-time corresponds to carrier localization in local weakly localized states, which is more quenched by non-radiative recombination and competes with carrier transporting to local strongly localized state. A proof of this is that the brightest place (point A) does not have fast decay, since the bandgap energy reaches its minimum at point A and carriers are most strongly localized, with barely any escaping. On the other hand, the slow decay with large life-time corresponds to carrier localization in local strongly localized state.

Comparing the life-time data in last column in Table I, it is notice that from bright region to dark region, the slow decay life-time becomes smaller. As mentioned before, the bandgap energy in bright region is smaller than that in dark region. So from point A to point H, with the decrease of the PL intensity, the bandgap energy increases, carriers are less localized and it’s easier for them to escape to non-radiative recombination centers, which increases the probability/rate of non-radiative recombination. As a result, from bright region to dark region, non-radiative recombination life-time decreases and slow decay life-time decreases.

Table I also shows that the fast decay life-time increases from bright region to dark region. However, from previous analysis it is expected to see that similar as $\tau_2$, $\tau_1$ also increases with increased PL intensity. Since $\tau_1$ is related with both carrier transportation and carrier non-radiative recombination, it is believed that the carrier transportation plays an important role. In order to have increased $\tau_1$ from bright region to dark region, the transportation rate of carriers transporting from local weakly localized state to local strongly localized states should be larger in bright region than in dark region.

Conclusions

In summary, temperature-dependent PL and microscopic TRPL were used to analyze the carrier localization and carrier transportation in blue-emitting InGaN/GaN MQWs. In temperature-dependent PL study, the change of integrated-PL intensity was fit with Arrhenius equation and explained with carrier localization in two localized states; the S-shaped PL peak energy and inverse-S-shaped PL FWHM with increasing temperature was explained with carrier localization, carrier transportation and shift in carrier life-time. In TRPL study, two lifetimes were observed in almost any place except the place with highest PL intensity. It was observed that the fast decay life-time increases with decreasing PL intensity, indicating a higher carrier transportation rate at bright region, while the slow decay life-time decreases at reduced PL intensity, indicating a higher probability of non-radiative recombination at dark region.

Acknowledgments

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