Carrier localization and nonradiative recombination in yellow emitting InGaN quantum wells

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InGaN quantum wells, with luminescence in the yellow region of the visible spectrum, have been studied using conventional and time-resolved cathodoluminescence. We observe the absence of strong localization effects and a relatively high internal quantum efficiency of ~12%, which are unexpected for InGaN in this long wavelength emission range. We have also observed a steady decrease of the peak emission energy, and a continuous increase in the radiative recombination lifetime with temperature up to 100 K. These two features are manifestations of recombination due to nonlocalized excitons. Nonradiative recombination centers, with activation energy of ~6 meV, appear to constitute the main mechanism limiting the internal quantum efficiency of these films.


InGaN quantum wells (QWs) grown along the polar c-axis of GaN are widely used in blue and green light emitting diodes (LEDs). Their distinct piezoelectric properties and excitonic nature have drawn considerable attention. In spite of the high concentration of structural defects, their internal quantum efficiency is surprisingly high. This is possibly due to the spatial localization of excitons associated with compositional inhomogeneities. One indication of carrier localization is the so-called S-shaped temperature dependence of the luminescence peak energy, characterized by an initial redshift with temperature \(T < T_1\) followed by a blueshift \(T_1 < T < T_2\) and finally another redshift at higher temperatures \(T > T_2\). The characteristic temperatures \(T_1\) and \(T_2\) vary significantly for QWs emitting in different spectral regions. For ultraviolet LEDs, a blueshift is observed between \(T_1 = 10\) K and \(T_2 = 20\) K, while for blue LEDs a continuous blueshift is observed beyond room temperature \(T > 300\) K. Such energy shifts with temperature can be explained in terms of an exciton thermalization and relaxation model. While excitons may be immobile at very low temperatures, for \(T > T_1\) they are able to overcome the potential fluctuations and relax into lower potential minima. This becomes evident by a redshift in the emission peak. In the temperatures range \(T_1 < T < T_2\), delocalization of excitons out of potential minima may occur, which results in a blueshift of the emission peak. For temperatures higher than \(T_2\), a decreasing value of the band gap energy results in the Varshni redshift of the emission energy. The intermediate blueshift in the S-shaped behavior is considered evidence of the presence of localized states. The respective temperature ranges reflect the degree of potential energy fluctuation.

The second important factor is the dimensionality of the exciton in its various stages of localization. Excitons confined in a zero-dimensional potential (e.g., a quantum dot) exhibit a radiative lifetime that is almost constant with temperature. For excitons in two-dimensional (2D) extended states (e.g., a QW), the radiative lifetime increases proportional to temperature. Thus, the temperature dependence of the radiative lifetime provides a direct measure of the exciton dimensionality. Monochromatic cathodoluminescence (CL) images of In\(_{0.1}\)Ga\(_{0.9}\)N QWs can reveal spatially distributed localization centers with dimensions under 60 nm, with a resolution limited by the carrier diffusion length. Such localization centers are usually assigned to indium-rich regions acting like quantum dots.

In this letter, we report the effects of carrier localization and nonradiative recombination on the optical properties of InGaN QWs that unintentionally exhibit a very long wavelength emission at ~570 nm in the yellow spectral region. We report the absence of apparent localization based on the temperature dependence of the emission energy and the radiative lifetimes. Nonradiative recombination is found to be the dominant recombination mechanism at room temperature.

The InGaNGaN thin film structure was grown by metalorganic vapor phase epitaxy. The active region consists of five InGaN QWs with a thickness of ~3 nm, separated by GaN barriers 15 nm thick, and capped with a GaN layer 100 nm thick. As intended, the structure should not be any different from samples emitting in the 525–545 nm range. CL spectra were obtained for temperatures ranging from 5 to 300 K, in a scanning electron microscope operated at an electron acceleration voltage of 5 kV with a beam current of 400 pA. Time-resolved CL (TRCL) was performed with a 100 ns electron pulse width and a 1 MHz repetition rate. The pulse rise and fall times were below 100 ps with an overall system resolution under 400 ps measured with a GaAs photocathode.

The microstructure of the InGaN QW region is shown in the transmission electron microscope (TEM) images in Fig. 1. The contrast in these multibeam high-resolution TEM images corresponds to the difference in indium compositions in the layers. The QW contrast is inhomogeneous, with islands of lateral dimensions between 15 and 30 nm [Fig. 1(b)]. We observe a dark line in the center and a fading contrast toward the GaN barriers. Small clusters of 2 to 3 nm in diameter are observed in the high-resolution image of Fig. 1(c). The clusters that are well oriented along the projection exhibit a
spherical shape. Considering that the TEM image is a 2D projection along the [110] axis, the spatial distribution and orientation of the indium cluster units along the zone axis may result in this symmetric contrast since they are much smaller than the specimen thickness (~100 nm). Limitations from TEM sample preparation and possible instability of very thin InGaN samples prevent us from making a stronger statement about the nature of these films. Nevertheless, these structural characteristics may correlate with the surprisingly long emission wavelength, and differ from other thin film structures emitting in the 530–545 nm range.\textsuperscript{13}

The variation in the InGaN QW emission spectra in the temperature range from 5 to 300 K is shown in Fig. 2. The peak energy shifts from 2.24 eV (553 nm) at 5 K to 2.15 eV (577 nm) at 300 K. The linewidths of the emission peaks are \textasciitilde 195 meV, and do not vary significantly with temperature. These are large values compared to 80 meV for blue and green emitting QWs, and may be due to compositional disorder. The emission peaks in the spectra were closely fit with Gaussian functions, with a standard deviation of $E = 0.001$ eV. The peak energy obtained from such fittings, plotted in Fig. 2(b), decreases monotonically with temperature. No S-shaped variation with temperature is observed; or more specifically, we observe no blueshift associated with a transition from localized into extended states. These results suggest the absence of significant carrier localization effects on the emission properties of the QWs. A possible explanation is that in higher indium content QWs, indium clusters acting as localization centers are no longer electronically isolated. They are sufficiently small, so that the states in the respective potential minima overlap and form a miniband. Carriers then occupy extended states in the miniband, and are no longer spatially localized. The emission energy is due to transition between minibands in the conduction and valence bands. As a result, no blueshift related to the transition from localized states to extended states is observed.

In order to further understand the carrier dynamics, TRCL was performed as a function of temperature. Figure 3 shows luminescence transients taken at the QW emission peak at 5, 100, and 300 K. After the electron beam is switched on at $t=0$ ns, the sampled region gets optically excited and the CL intensity builds up. This behavior is similar for all temperatures. At $t=100$ ns, the electron beam is switched off and the luminescence decay is recorded. At 5 K, the luminescence decay is very fast, probably dominated by the radiative process. With increasing temperature, for example, at 100 K, the decay becomes much slower. This can be explained in terms of a competition between radiative and nonradiative recombination paths, since nonradiative centers are thermally activated with temperature.\textsuperscript{14} However, at temperatures above 100 K, the CL decay remains almost independent of temperature, perhaps due to saturation of the nonradiative recombination.

The internal quantum efficiency of the QW can be estimated from the temperature dependence of the luminescence intensity. Figure 4(a) shows the wavelength-integrated luminescence intensity (represented by dots) in the temperature range from 5 to 300 K. An upper value of the internal

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**FIG. 1.** Transmission electron microscopy images of a yellow-emitting QW. (a) Dark field image with $g=(0002)$. (b) High resolution image of all five QWs and (c) high resolution image of top two QWs.

**FIG. 2.** (Color online) CL of the QW measured in the temperature range from 5 to 300 K. (a) Normalized spectra taken at temperatures specified on the right. (b) Plot of the QW emission peak energy vs temperature.

**FIG. 3.** (Color online) TRCL transients for the peak emission energy at 5, 100, and 300 K. The electron beam is switched on at $t=0$ ns and off at $t=100$ ns, with a repetition rate of 1 MHz.

**FIG. 4.** (Color online) (a) Wavelength-integrated luminescence intensity plotted for InGaN QWs at a temperature range from 5 to 300 K.
quantum efficiency ($\eta$) can be determined assuming that radiative recombination is dominant at sufficiently low temperatures.\cite{15,16} The temperature dependence of the CL lifetime ($\tau$), the radiative lifetime ($\tau_R$), and the nonradiative lifetime ($\tau_{NR}$) is shown in Fig. 3(b). The CL lifetimes were obtained by fitting the transients with an exponential decay function. The radiative and nonradiative lifetimes were derived on the basis of the following equations:

$$\eta = \frac{1}{\tau_R} \left( \frac{1}{\tau} \right)$$

(1)

$$\frac{1}{\tau} = \frac{1}{\tau_R} + \frac{1}{\tau_{NR}}$$

(2)

As seen in Fig. 4(b), the radiative lifetime increases proportionally to temperature up to \( \sim 100 \) K. This linear behavior is characteristic of 2D exciton recombination inside a QW. There is no temperature range where the radiative lifetime remains constant, an indication of localized excitons. This is further support for exciton localization not playing a major role in this material. The absence of localization effects agrees with an earlier report on high-power green LEDs by some of us.\cite{17} The nonradiative recombination lifetimes decrease rapidly with temperature, and become smaller than the radiative lifetime above 100 K. The thermal activation energy for the nonradiative recombination process can be extracted from the temperature dependence of the emission intensity in Fig. 3(a). The data were fitted using the following expression:

$$I(T) = \frac{I_0}{1 + C \exp \left( \frac{E_A}{k_B T} \right)}$$

(3)

The activation energy was found to be 6 meV, which is a very small potential depth for carrier localization. This may imply that the major limitation of the quantum efficiency at room temperature is due to the nonradiative recombination, possibly at the cluster boundaries. The reduction in nonradiative centers could contribute to high efficiency more than the indium composition inhomogeneities.

In summary, we have investigated carrier localization and the effects of nonradiative centers in InGaN QWs emitting in the yellow range of the visible spectrum. From the temperature dependence of CL spectra, we show that the peak energy exhibits no blueshift, which would be related to thermalization of localized excitons to extended states. The temperature dependent TRCL measurement confirms the absence of pronounced effects due to carrier localization. The radiative lifetime increases proportionally with temperature, which indicates a 2D behavior rather than zero-dimensional localization. The existence of nonradiative recombination centers with low thermal activation energies is the limiting factor of the internal quantum efficiency. These may be related to the small cluster boundaries in the QWs.

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