Multiple-component photoluminescence decay caused by carrier transport in InGaN/GaN multiple quantum wells with indium aggregation structures

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(Received 7 January 2002; accepted for publication 13 April 2002)

Based on wavelength-dependent and temperature-varying time-resolved photoluminescence (PL) measurements, the mechanism of carrier transport among different levels of localized states (spatially distributed) in an InGaN/GaN quantum well structure was proposed for interpreting the early-stage fast decay, delayed slow rise, and extended slow decay of PL intensity. The process of carrier transport was enhanced with a certain amount of thermal energy for overcoming potential barriers between spatially distributed potential minimums. With carrier supply in the carrier transport process, the extended PL decay time at wavelengths corresponding to deeply localized states can be as large as 80 ns. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1484546]

Because of the large lattice mismatch between InN and GaN, their low miscibility leads to indium aggregation and phase separation.[1,2] Such results of spinodal decomposition produce quantum-dotlike structures in InGaN/GaN quantum wells (QWs). The cluster structures form spatial potential fluctuations and localized energy states for trapping carriers.[3,4] Due to carrier localization, time-resolved photoluminescence (TRPL) measurements have shown quite long photoluminescence (PL) decay times.[5] In particular, multiple-component decays of TRPL have been observed.[5–7] Although a nonsingle-exponential model with a β parameter was used for fitting such multiple-component decay processes,[6,7] this model did not provide much physical insight. Actually, the physical meanings of the multiple-component PL intensity decay have not been well discussed yet. In this letter, based on wavelength-dependent and temperature-varying TRPL measurements, we propose the mechanism of carrier transport between different levels of localized states, which are spatially distributed, for interpreting the observed multiple-component PL decay. The difference in decay time between the early-stage fast decay and the extended slow decay is mainly attributed to carrier transport from certain localized states into other states of stronger localization. Also, the long single-exponential decay of PL intensity at the photon energies, corresponding to deeply localized states, is supposed to be due to carrier supply in the carrier transport process.

The sample used in this study was grown in a low-pressure metalorganic chemical vapor deposition reactor. The InGaN/GaN multiple QW sample consisted of five periods of Si-doped InGaN well with 3 nm in thickness. The designated indium composition was 16%. The Si doping concentration was 10^{18} cm^{-3}. The barrier was 7 nm GaN. In the sample, the QW layers were sandwiched with a 1.5 μm GaN buffer layer on a sapphire substrate and a 50 nm GaN cap layer. The growth temperatures were 1050 °C and 740 °C for GaN and InGaN, respectively. The TRPL measurements were performed using a Hamamatsu streak camera with time resolution about 5 ps. Frequency-doubled optical pulses with 100 fs pulse width and 76 MHz pulse repetition frequency were generated from a mode-locked Ti:Sapphire laser for excitation. The excitation photon energy and power of TRPL were 3.1 eV (400 nm) and 60 mW, respectively.

Figure 1 shows time evolution of PL spectrum at 12 K. One can see that the spectral intensity decays slower on the low-energy side, when compared with that on the high-energy side. Also, as shown in the inset of Fig. 1, the PL peak position redshifts fast in the early stage and then slows down. The fast shift in the early stage may imply the process of carrier transport. Low-temperature (12 K) temporal variations of PL intensity at several emission photon energies are

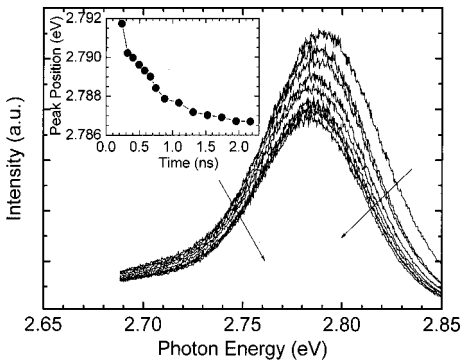

FIG. 1. Time evolution of PL spectrum at 12 K. The inset shows temporal variation of the photon energy at the PL spectral peak.

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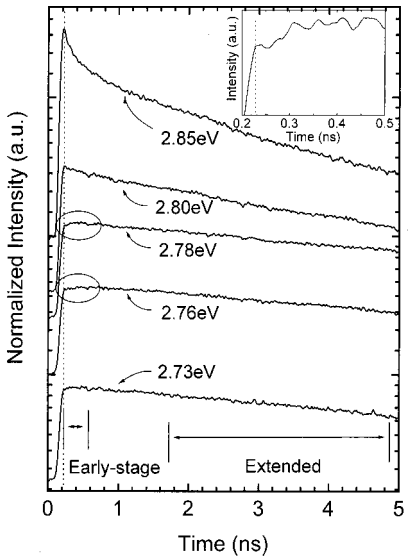

FIG. 2. Temporal profiles of PL intensity at several photon energies when sample temperature is 12 K. The inset shows the delayed slow rise on a finer scale.

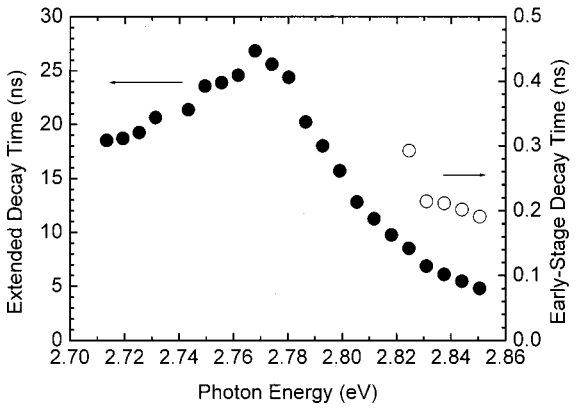

FIG. 3. Calibrated early-stage decay times (the right ordinate) and extended decay times (the left-hand side ordinate) as functions of photon energy at 12 K.

shown in Fig. 2. Because we used excitation pulses of 76 MHz pulse repetition frequency, within one excitation cycle (13.6 ns) PL does not completely disappear. This fact explains the different initial PL intensity levels of various photon energies, as can be seen at the left-hand side end of Fig. 2 (also in Fig. 4). In Fig. 2, the curves corresponding to the high-energy side (the curves of 2.85 and 2.80 eV) show multiple-exponential decays, which are relatively faster in the early stage and slower in the extended range. The fast-decay behavior disappears when emission photon energy is below a certain value around 2.8 eV. Near PL spectral peak, in the early stage, there exists a delayed slow rise for a few hundred ps, as highlighted with the ellipses in the curves of 2.78 and 2.76 eV. The inset in Fig. 2 shows the delayed slow rise on a finer scale in the curve of 2.78 eV. These dull shapes of temporal behavior are particularly clear when they are compared with the curves of 2.85 and 2.8 eV. Such behaviors become weaker in the PL intensity profiles on the low-energy side, as indicated by the curve of 2.73 eV in Fig. 2. A similar temporal behavior of delayed rise was observed in a GaN/AlGaN double heterostructure.[8] It was claimed that this delayed rise behavior of GaN was due to the effect of carrier accumulation with a capture process from the AlGaN cladding layer into the GaN active region. In the InGaN/GaN system, due to spatial fluctuation of a potential level, carriers may transport among different levels of localized states. Thus, we propose that this delayed slow rise be essentially due to the carrier capture process from weaker localized states (higher potential minimums) into stronger localized states. The carrier capture process can also be used for interpreting the early-stage fast decay on the high-energy side (curves of 2.85 and 2.8 eV). Because of fast carrier outflow, PL intensity decays fast on the high-energy side.

On the high-energy side, we fitted the multiexponential curves with the biexponential model to obtain the early-stage and extended decay times.[9,10] On the other hand, a single-component decay profile was fitted with a single-exponential curve (except the delayed slow rise duration). Note that the stretched exponential model with the parameter β (Ref. 6) can not fit the early stages of the decay profiles well on the high-energy side. Figure 3 shows the emission photon energy dependent decay times (different from carrier lifetimes) at 12 K. On the high-energy side, the fast decay time can be shorter than 200 ps. Such a short decay time at 12 K can not be attributed only to radiative recombination, either free carrier recombination or exciton recombination. Besides recombination, an additional process, i.e., carrier transport from weakly localized states to strongly localized states, must be considered. The decreasing trend of the early-stage fast decay time with increasing photon energy, as shown with the empty symbols in Fig. 3, is reasonable because of the faster carrier escape from weaker localized states.

As for the extended decay component, the decay time increases first and then decreases with increasing emission photon energy. The decreasing trend right to the peak is attributed to weaker carrier capture of relatively shallower localized states in the extended time range. It can also be partially due to faster recombination in weaker localized states. The shorter decay times at the low-energy ends of spectra, compared with those of energy levels near the PL peak, can be explained with the fact that carrier transport into strongly localized states may require a certain energy to overcome a potential barrier. In other words, it is more difficult for carriers to transfer into strongly localized states. This argument is consistent with the absence of the delayed slow rise behavior in PL intensity evolution on the low-energy side, as indicated by the comparison between the curves of 2.78 and 2.73 eV in Fig. 2. The existence of the delayed slow rise behaviors implies easier carrier capture of those states near the PL peak.

Figure 4 shows PL intensity decay profiles on the low-energy side (below PL peak by 43 meV) of the sample at several temperatures. Below about 150 K, the delayed slow rise can be observed (as again highlighted with the ellipses) within the first 1 ns evolution. This behavior becomes more prominent as temperature increases. After this rise, PL intensity shows slow extended decay, indicating long-term carrier capture of deeply localized states. When temperature increases up to 180 K, clear two-component decays can be observed. The fast early-stage decay lasts for less than 1 ns. The difference between the fast early-stage decay and the slow extended decay at a relatively higher temperature is

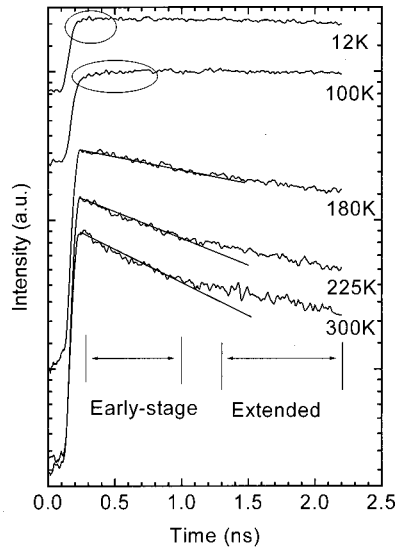

FIG. 4. Temporal profiles of PL intensity at several temperatures when photon energy is below the peak by 43 meV. The ellipses highlight the delayed slow rises of PL intensity.

attributed to carrier escape. Significant carrier escape only at a higher temperature reveals the result of thermal energy enhanced carrier dynamics. The fast-decay is caused by thermal carrier escape into either higher or even lower levels. The escape is more prominent as temperature increases. Such a trend is clearly shown with the empty squares in Fig. 5. In Fig. 5, the fast early-stage decay times of the sample at the PL peak photon energy, a photon energy on the high-energy side (above the peak by 64 meV), and a photon energy on the low-energy side (below the peak by 43 meV) are represented by the empty circles, triangles, and squares, respectively. One can see that as photon energy increases, the temperature for observing multiple-component decay or fast early-stage decay becomes lower. On the high-energy side, the fast early-stage decay, or carrier escape occurs at the lowest temperature and higher than our measurement. However, near the PL peak, the carrier escape behavior does not occur until the temperature rises to about 100 K. Thermal energy helps

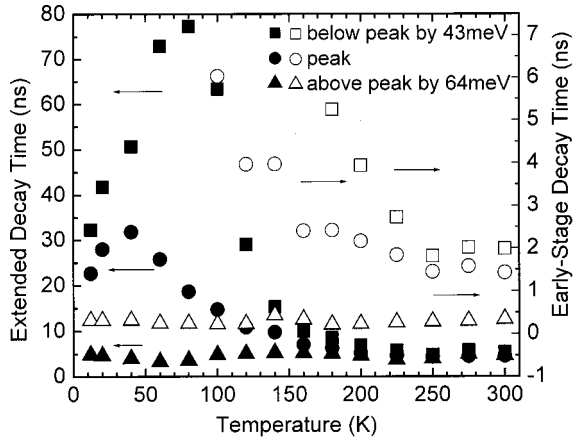

FIG. 5. Calibrated early-stage decay times (the right ordinate) and extended decay times (the left-hand side ordinate) as functions of sample temperature for three photon energies.

in overcoming the potential barrier for carrier escape from relatively stronger localized states.

Figure 5 also shows the extended decay times at the three photon energies, as indicated by the filled symbols. One can see that for photon energies near the PL peak and on the low-energy side, the decay times increase first and then decrease as temperature rises. The longer decay times on the low-energy side are attributed to better carrier localization (from nonradiative recombination) and extended carrier capture. The decreasing trend at relative higher temperatures is due to stronger nonradiative recombination and possibly carrier escape. The increasing trend in the low-temperature portion can be attributed to the enhanced carrier transport process with a reasonable amount of thermal energy. However, with thermal energy exceeding a certain level, corresponding to the peak of filled symbol distribution, enhanced nonradiative recombination dominates carrier dynamics and PL decay times at photon energies near the PL peak or on the low-energy side decrease with increasing temperature. This critical thermal energy level increases with decreasing photon energy because nonradiative recombination is less effective for carriers in strongly localized states. The temperature-dependent behavior of the extended decay time on the high-energy side is quite different. There is a minimum around 60 K. This behavior can be attributed to the enhanced carrier escape from weakly localized states with a reasonable amount of thermal energy before nonradiative recombination dominates.

In summary, we have shown the data of wavelength-dependent and temperature-varying TRPL measurements in an InGaN/GaN QW sample. The multicomponent PL intensity decay behaviors were interpreted as the results of carrier transport from relatively weaker localized states into stronger localized states. The process of carrier transport was enhanced with a certain amount of thermal energy.

This research was supported by National Science Council, The Republic of China under the grants of NSC 90-2112-M-002-052, NSC 90-2215-E-002-027, and NSC 90-2215-E-002-041.

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