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Investigation of fast and slow decays in InGaN/GaN quantum wells

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We have measured and analyzed the photoluminescence spectra from InGaN/GaN multiple quantum wells. Emission peaks due to recombination of the photogenerated carriers occupying localized states and extended states within quantum wells have been identified through temperature-dependent photoluminescence. Fast and slow decays have been attributed to recombination of carriers in extended states and localized states, respectively, based on time-resolved pump-probe differential photoluminescence. © *2011 American Institute of Physics*. [doi:10.1063/1.3627166]

Understanding the emission mechanism in InGaN/GaN quantum wells (QWs) is the key for the applications in light-emitting devices from ultraviolet to green region.[1] Previously, recombination from localized states was proposed as the primary mechanism for spontaneous emission in InGaN/GaN QWs.[2] Such an effect of carrier localization has been attributed to nanometer-scale indium rich clusters based on transmission electron microscopy until these indium clusters were attributed to the electron-beam-induced damages.[3] Besides indium rich clustering effect, v-shaped pits can induce carrier localization.[4] Recently, it was demonstrated that InGaN QWs can be grown with no indium clustering and abrupt interfaces.[5] In addition, polarization fields cause charge separation and, therefore, affect the carrier lifetime due to different recombination mechanisms.[6–9]

Time-resolved photoluminescence (PL) was widely used to investigate the dynamic recombination processes in InGaN/GaN QWs. Carrier lifetimes in the range from sub-nanosecond to microsecond have been measured, which suggests that the carrier lifetime can be greatly influenced by the width of QWs and indium concentration.[6,7,10,11] Specifically, two-step PL decay was observed with the early fast and later slow decay processes being attributed to carrier transfer from weakly to strongly localized states and recombination of carriers in strong localized states, respectively.[12]

In this letter, using time-resolved pump-probe differential PL,[13] we have directly measured the fast decay time constants of the photogenerated carriers in the range of 1.41-2.22 ns, which is attributed to the lifetime of carriers in QWs. Moreover, the time constant of extremely slow decay of dominant PL peak is identified as long as 570 ns, corresponding to the recombination of the carriers at localized states.

In our studies, the sample, grown by metal-organic chemical vapor deposition, consists of four periods of InGaN/GaN QWs with 3 nm $In_{0.2}Ga_{0.8}N$ QWs sandwiched by 12-nm thick GaN barriers, respectively. The growth method and device structure of this sample are similar to those outlined in Ref. 14. A laser beam generated by Ti:sapphire regeneration amplifier with a pulse width of 180 fs, and a repetition frequency of 250 kHz was frequency-doubled by a β-barium borate (BBO) nonlinear crystal. As a result, such an amplifier system produced a train of the pulses at 393 nm. The radiation beam was split into two parts labeled by us as pump and probe, having relatively high and low powers, respectively. The probe beam was passed through a chopper, whereas the pump beam was sent through a delay line to vary the temporal delay for probe pulses relative to pump pulses. Since the probe beam was modulated, one can measure the PL intensity generated by the probe beam through locking into the modulated frequency via a lock-in amplifier. The PL signal is focused into a spectrometer and then detected by a photomultiplier tube.

Fig. 1(a) illustrates PL spectra measured at 5 K under different excitation power densities. The main peak (P_L) is located at 2.575 eV at an excitation power density of 0.5 W/cm^2. The multiple peaks on the low energy side are LO-phonon replica.[15] When the excitation intensity is increased, a shoulder (P_H) on the high energy side appears, which suggests the presence of one peak or multiple ones. Multiple peaks in InGaN/GaN were widely observed and attributed to splitting of valence band,[16] different quantum states,[17] and weak-strong localized states.[18] Using one-dimensional Schrödinger-Poisson solver under the carrier density of 5.4×10^{12} cm^{-2} at the pump intensity of 30 W/cm^2, the energy of the lowest QW transition (i.e., e1-hh1) is calculated to be 2.741 eV at 5 K. This calculated transition energy is higher than that of the dominant peak in Fig. 1(a) by about 140 meV. In comparison, it is only lower than that of the broad shoulder on the high-energy side by about 10 meV. As analyzed below, the main peak and shoulder are caused by the recombination of the carriers at localized states and extended states, respectively.[19]

Fig. 1(b) shows the behavior of emission energy of P_L versus excitation density. A blue shift in the amount of 30 meV is observed when the excitation density is increased from 170 mW/cm^2 to 16.7 W/cm^2. As excitation density is further increased up to 3200 W/cm^2, the P_L exhibits a red shift in the amount as large as 49 meV. Previously, the blue shift was frequently observed and explained by the

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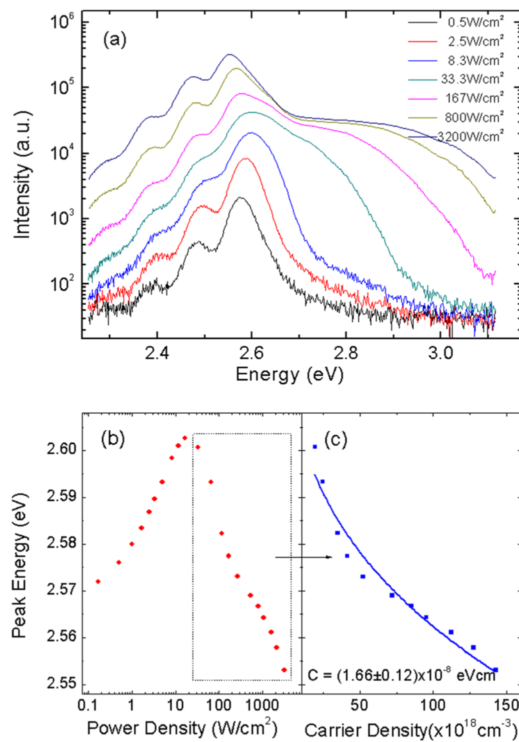

FIG. 1. (Color online) (a) Excitation power density dependent PL spectra for InGaN QWs measured at 5 K. (b) The emission energy of P_L vs excitation power density measure at 5 K. (c) The emission energy of P_L vs carrier density for excitation power density higher than 16.7 W/cm^2. The solid curve is a fit based on band gap renormalization effect.

combination of band-tail filling effect and reverse quantum confine Stark effect (QCSE) as the carrier density in QWs is increased.[20–22] However, the anomalously large red shift has not been observed yet in InGaN/GaN QWs. To understand the origin for such red shift, we plot the peak energy of P_L (only red shift part) versus carrier density, as shown in Fig. 1(c). Since the thickness of QWs is much less than a typical absorption depth of the excitation, we can assume spatially homogeneous excitation of well layers. In such case, the photogenerated carrier density can be determined as $n = \alpha J/(h\nu S)$, where α is the absorption coefficient, J is the energy per pulse, $h\nu$ is the incident photon energy, and S is the focus area of incident laser. Under very high laser intensities, the absorption efficient could be reduced due to band-filling effect. Thus, we calculate the carrier intensity at high excitation powers by $n = n_0 I/I_0$, where I and I_0 are integrated PL signals at high and low excitation powers, respectively. The peak energy as a function of carrier density can be well fit via an expression of band-renormalization effect, $E = E_0 - Cn^{1/3}$, see Fig. 1(c). The fitting value of C is obtained as 1.66×10^{-8} eV cm, which is very close to that in Ref. 22. Therefore, such a red shift is likely the manifestation of band-gap renormalization.

The peak energy of P_L is plotted as a function of temperature measured at 16.7 W/cm^2, see Fig. 2(a). A blue shift as large as 32.4 meV from 5 K to 120 K has been observed. The increase of the peak energy with temperature in InGaN QWs is considered as an indication of emission from localized states.[23] Such temperature-dependent emission energy at localized states can be described as

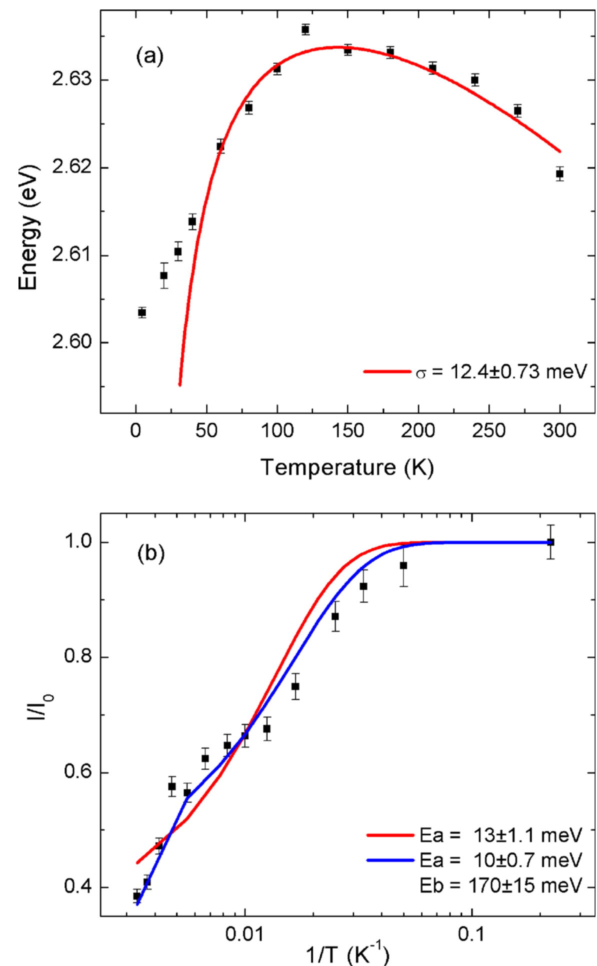

FIG. 2. (Color online) (a) The emission energy of P_L vs temperature measured at 16.7 W/cm^2. The solid curve is a fit based on band tail model. (b) Normalized integrated PL intensity as s function of $1/T$ for InGaN QWs measured at 16.7 W/cm^2. The solid curves are single and two channels Arrhenius fits, respectively. E_a and E_b stand for the activation energy.

$$E(T) = E(0) - \frac{\alpha T^2}{T + \beta} - \frac{\sigma^2}{k_B T},$$

where $E(0)$ is the energy gap at zero temperature, α and β are the Varshini's parameters, σ indicates the degree of the localization effect, and k_B is the Boltzmann constant. Using this formula to fit the data, see Fig. 2(a), we obtained σ to be 12.4 meV, which is consistent with that in Ref. 24. Therefore, we have attributed the main peak in PL spectra to the recombination of carriers occupying localized states. To further verify the carrier localization, both single and two-channel Arrhenius equations[25] were used to fit temperature-dependent PL intensities, see solid curves of Fig. 2(b), respectively. The activation energy is determined to be 13 meV in the single-channel fit which is quite close to the value of σ. Since this activation energy is much less than the QW band offsets, the thermal quenching originates from thermionic emission of carriers out of potential minima caused by localized states rather than the thermal activation of electrons and holes out of InGaN QWs. The two-channel Arrhenius equation provides a better fit, especially at high temperature, which could be caused by the increase in the intensity of P_H relative to that of P_L with increasing temperature. The activation energies obtained from such a fit are

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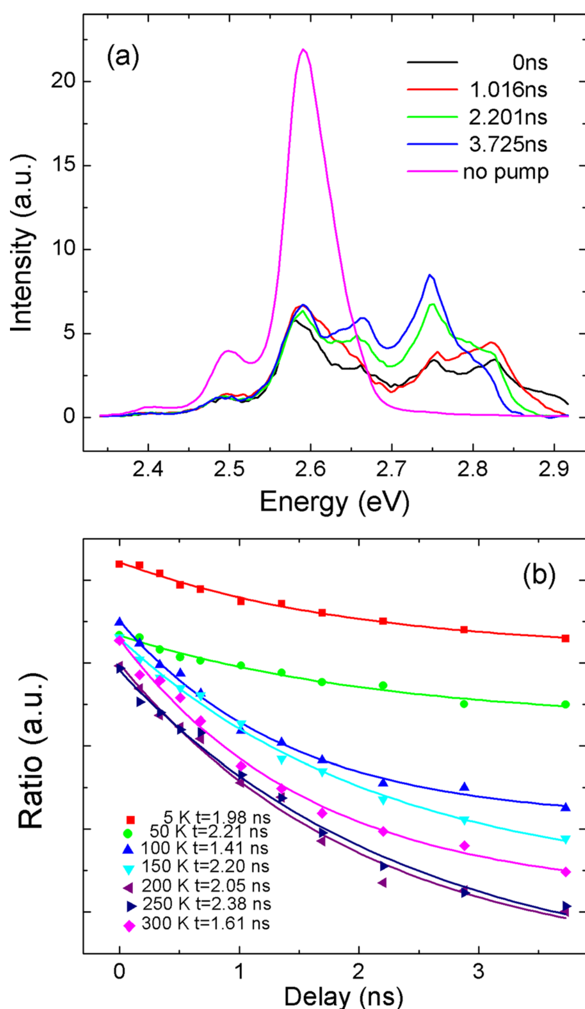

FIG. 3. (Color online) (a) Pump-probe differential PL spectra at different delay times measured at 5 K. (b) Differential ratio vs. delay time measured at different temperatures. The solid curves are single exponential fits to experiment data.

11 meV and 169 meV, respectively. We tentatively attribute these energies to carrier confinement energy of localized states and thermal escape energy for the carriers moving out of QWs, respectively.

We have measured the PL spectra at different delay times for the pump beam, see Fig. 3(a). When the pump is turned on, the intensity of P_L is reduced dramatically, whereas that of P_H increases. The intensity of P_L does not recover even if we increase the delay time to 3.725 ns, which suggests that P_L has a very long lifetime, i.e., slow decay. Using a fast oscilloscope, we have measured the slow decay time constant of 570 ns, corresponding to lifetime of the carriers occupying localized states. On the other hand, the intensity of P_H increases obviously when the delay time is increased, which implies that P_H has a fast decay. Fig. 3(b) is a plot of the differential ratio vs. delay time. The differential ratio can be expressed by $R = 1 - I(t)/I_0$, where I_0 is the integrated PL intensity induced by the probe without the pump and $I(t)$ stands for that of the probe with the pump at a certain delay instant t. Considering that the slow decay has a much longer lifetime compared with the fast decay, a single exponential decay function is used to obtain the fast decay time as a function of the temperature. As a result, the decay time constants are determined to be 1.41-2.22 ns. Obviously, the fast decay time is more or less independent of the temperature, which indicates that nonradiative recombination plays a minor role.

In conclusion, the emission peaks from localized states and extended states have been observed in the InGaN/GaN QWs. By analyzing the dependence of the PL spectra on the excitation power, a red shift has been observed, due to bandgap renormalization. Moreover, through pump probe differential PL measurements, we confirm that the fast decay originates from the lifetime of the carriers in extended states, whereas the slow decay is the manifestation of the recombination of the carriers at localized states.

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