Hole escape processes detrimental to photoluminescence efficiency in a blue InGaN multiple-quantum-well diode under reverse bias conditions

T. Inoue and K. Fujiwara^{a)} Kyushu Institute of Technology, Tobata, Kitakyushu 804-8550, Japan

J. K. Sheu

Institute of Electro-Optical Science and Engineering, National Cheng-Kung University, Tainan, Taiwan 70101, Republic of China

(Received 1 February 2007; accepted 16 March 2007; published online 17 April 2007)

Photoluminescence (PL) properties of a blue $In_{0.3}Ga_{0.7}N$ multiple-quantum-well (MQW) diode with an additional n^+ -doped $In_{0.18}Ga_{0.82}N$ electron reservoir layer (ERL) have been investigated at 20 K as a function of reverse bias under indirect barrier excitation. A PL intensity ratio of MQW/ERL is observed to be significantly quenched by increasing the reverse field due to electron-hole separation and carrier escape, in spite of observed blueshifts, when the excitation power is decreased by two orders of magnitude. The PL intensity reduction suggests that the hole escape process plays an important role for determination of the PL efficiency under the reverse bias. © 2007 American Institute of Physics. [DOI: 10.1063/1.2723683]

Despite the realization of blue and green light-emitting diodes (LEDs) based on InGaN/GaN quantum-well (QW) heterostructures,^{1,2} the origin of the very bright emission characteristics is still controversially discussed.^{3–8} A peculiar property of this material system is the observation of efficient luminescence, although the density of misfit dislocations can be as high as 10^{10} cm⁻². Therefore, we expect the existence of a particularly important mechanism, which is responsible for the enhancement of the radiative efficiency in the presence of a very high defect density. Previously quantum confinement effects on the InGaN alloy well and efficient carrier capturing by the localized radiative recombination centers have been claimed to be important for the origin of the high emission efficiency. Quite recently, importance of very efficient hole capture processes by localizing valence states associated with atomic condensates of In-N for radiative recombination efficiency is pointed out.^{9,10} Thus, all of the previous studies infer that carrier capture processes toward radiative recombination centers and prohibition of escape to nonradiative defective sites play an important role for the determination of the radiative recombination efficiency.3-13 In relation to assessment of the radiative recombination efficiency, we have recently investigated the temperature dependence of the electroluminescence (EL) intensity for a specially designed blue InGaN/GaN multiple-QW (MQW)-LED containing an additional n^+ -doped InGaN electron reservoir layer (ERL).^{14,15} This LED exhibits a significant improvement of the EL efficiency, in particular, for lower temperatures, when a forward bias necessary to obtain a certain injection current is high due to the reduced hole conductivity.

In this letter, photoluminescence (PL) properties of the blue InGaN MQW-LED with ERL have been investigated with a special emphasis on external field effects on the radiative recombination processes. The existence of n^+ -type ERL below the active MQW layer allows us to monitor how the photogenerated carrier distribution across the active MQW region influences the PL efficiency by changing exci-

tation power as a function of field strength. Observed PL intensity reduction induced by the reverse fields suggests importance of hole escape processes from the MQW for the determination of the PL efficiency.

An InGaN/GaN MQW-LED with an additional n^+ -doped In_{0.18}Ga_{0.82}N ERL was grown by metal-organic vapor-phase epitaxy.¹⁶ The emission region of the LED consists of a triple In_{0.3}Ga_{0.7}N QW with a nominal width of 2.5 nm separated by 6.5 nm GaN barriers. Details of the MQW-LED heterostructure were described previously.^{15,16} PL spectra have been recorded over a wide spectral range as a function of forward and reverse bias voltages at 20 K with a lock-in detection technique, using a He–Cd laser at 325 nm for indirect photoexcitation at various excitation powers of 0.1–10 mW (power density of ~1–10² W/cm²).

Figure 1(a) shows PL spectra of the diode taken with a 10 mW laser power and at 4.25, 2.0, 0, and -3.0 V. When excited from the surface p-GaN cap layer, the MQW diode shows a main blue MQW emission band around 480 nm, which is strongly redshifted due to carrier localization from absorption band tails, as confirmed by photocurrent spectra (not shown). In addition to the main blue emission band, a distinct PL band at 405 nm is observed only for the diode with ERL, but not for a similar MQW diode without it. Therefore, the PL band at 405 nm is identified as originating from the ERL located below the active MOW layer. A broad short-wavelength emission band is also observed around 380-440 nm, the origin of which is not clear at present. A small but sharp PL band observed at 355 nm is ascribed to bound excitons in the GaN layers. A broad PL band due to yellow emissions around 575 nm is also observed, only when the GaN barriers are indirectly photoexcited. When the forward bias is decreased and the reverse bias is increased to -3 V, the PL intensity of the main blue band is considerably decreased, accompanying blueshifts, while the emission intensity of the n^+ -doped ERL remains the same without any peak shifts. These results indicate that the external field is applied to the MQW region only and that the quantum confined Stark effect results in the compensation of the internal

90, 161109-1

^{a)}Electronic mail: fujiwara@ele.kyutech.ac.jp

^{© 2007} American Institute of Physics



FIG. 1. PL spectra of a blue $In_{0.3}Ga_{0.7}N$ MQW-LED with an additional *n*-type $In_{0.18}Ga_{0.82}N$ electron reservoir layer (ERL) as a function of forward (positive) and reverse (negative) bias voltages at 20 K under (a) intense (10 mW power) and (b) weak (0.1 mW power) indirect excitations at a wavelength of 325 nm. Note that the main PL peak around 480 nm shows a moderate (strong) intensity reduction in (a) [in (b)] with increasing the reverse bias voltage, accompanying blueshifts, while the ERL emission at 405 nm does not change its intensity. A small line seen at 650 nm is due to the laser scattering.

piezofield, which is opposite to the p-n junction field direction.^{5,13}

On the other hand, when the excitation power is decreased by two orders of magnitude to 0.1 mW, two substantial differences appear in the PL spectra, as shown in Fig. 1(b). That is, a PL intensity ratio of MQW/ERL is observed to be drastically increased at a forward bias of 4.25 V (near the flatband condition) due to a decrease of photon penetration depth (relative decrease of the ERL emission) and preferential photoexcitation of the front MQW region near the *p*-type clad layer. Secondly, the MQW PL intensity is significantly quenched by increasing the reverse field due to field-induced electron-hole separation and resultant carrier escape, in spite of observed blueshifts. Figure 2 shows normalized, wavelength-integrated PL intensity for MQW and ERL as a function of applied reverse bias at excitation powers of 0.1, 1.0, and 10 mW. It is clear that the PL intensity for MQW significantly decreases with increasing the reverse field and the reduction is stronger under the weak excitation power, while the ERL emission remains to be nearly constant. This field-induced PL intensity reduction dependent on the photoexcitation power suggests that the hole escape pro-



FIG. 2. Normalized, wavelength-integrated PL intensity for MQW and ERL bands as a function of reverse (negative) bias voltage at three different excitation powers of 0.1, 1.0, and 10 mW.

cess plays an important role for determination of the blue PL efficiency under the reverse bias conditions, as discussed in the following.

The bias dependence of the PL spectra significantly changes by decreasing the excitation power and the PL intensity decreases very rapidly with increasing the reverse bias (external field) under the weak excitation [see Fig. 1(b)]. Note that the PL intensity decreases to 18%, when the reverse bias is increased to -3 from +4.25 V. We attribute these PL spectral variations with decrease of the excitation power to the decreased excitation depth. This hypothesis can be easily confirmed because of the existence of the ERL. That is, the n^+ -type ERL which is located below the active region is more weakly photoexcited by the weak photoexcitation. We observe systematical decreases of the PL intensity for ERL relative to the main blue emission band, when the excitation power is decreased to 0.1 from 10 mW. But it is not surprising to find out that the ERL PL band does not show any discernible changes in intensity with bias at all excitation levels, since the ERL is heavily doped to n^+ type $(\sim 10^{19} \text{ cm}^{-3})$. However, when the reverse bias is increased, the intensity of the main PL band around 480 nm certainly decreases more slowly under the intense excitation, in contrast to the case of the weak excitation.

These variations of the PL intensity quenching by the reverse bias (reverse field) at various excitation powers can be explained in the following ways. Figure 3 illustrates the potential diagram of the MQW diode under the (a) weak and (b) intense excitation conditions. When the photon penetration depth is shallow as in Fig. 3(a), the photogenerated electron-hole pairs are not uniform across the MQW layer and more carriers are excited in the front well layer near the p-type barrier. Therefore, the photogenerated holes can escape more easily from the active well layer, when the reverse field is increased, leading to the rapid decreases of the PL intensity. However, under the intense excitation [see Fig. 3(b)] the MQW layer is rather uniformly excited and photoexcited carriers are generated deep into the bottom well layer. This is evidenced by the strong increase of the PL intensity for ERL. Therefore, the PL intensity of the main blue band results in slow decrease with increasing reverse bias, since holes generated in the bottom well near the ERL

Downloaded 19 Dec 2007 to 150.69.123.200. Redistribution subject to AIP license or copyright; see http://apl.aip.org/apl/copyright.jsp



FIG. 3. (Color online) (a) Under the weak excitation the photon penetration depth is shallow, so that the photoexcited holes from the active MQW regions can escape more easily, leading to the rapid decreases of the PL intensity with increasing the reverse bias. (b) Under the intense excitation the MQW layer is rather uniformly excited so that the main MQW band is more slowly decreasing with increasing the reverse bias, since photogenerated holes deep in the MQW regions need to traverse many barriers to escape from the active region.

must tunnel through the multiple barriers to escape to the p electrode. This result means that the hole escape process instead of the electron-hole wave function overlap changes is playing an important role in the radiative recombination efficiency in the active InGaN MQW layer under the reverse bias conditions. In other words, the PL emission efficiency under the reverse field is limited and reduced by the tunneling escape of holes, in spite of the fact that the QW potential is flattened as a result of the compensation of the piezoelectric field by the reverse field, as confirmed by Stark blueshifts.

Very recently, Chichibu *et al.*⁹ reported by studying positron annihilation experiments in InGaN alloys and QW layers that the hole capture processes by localizing valence states associated with atomic condensates of In–N play a very important role for the radiative recombination efficiency in InGaN materials. Our observation of the external field effects on the PL efficiency under the different photoexcitation depths may also indicate the importance of the hole capture (localization) processes in the active regions, since the radiative recombination efficiency is strongly modified by the hole escape ability under the presence of high density defects.

In summary, photoluminescence properties of a blue In-GaN MQW diode with a n^+ -type ERL have been investigated

as a function of bias voltage and excitation power. When the reverse bias is increased, the PL intensity of the main blue emission decreases due to external field-induced carrier escape from the radiative recombination centers within the wells, but the degree of reduction strongly depends on the excitation power because of the different photon penetration depths. Enhanced escape of photoexcited carriers under the weak photoexcitation suggests that hole escape processes play an important role in the radiative recombination efficiency in the active region of the diodes.

The authors would like to thank K. H. Ploog for helpful discussion on the importance of hole trapping for luminescence efficiency, H. Kostial and U. Jahn for sample die bonding and wiring, and N. Otsuji, H. Katou, and A. Satake for experimental assistance. This work was supported in part by the Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology (No. 16360157).

- ¹S. Nakamura and G. Fasol, *The Blue Laser Diode* (Springer, Berlin, 1997).
- ²I. Akasaki and H. Amano, Jpn. J. Appl. Phys., Part 1 36, 5393 (1997).
- ³S. Chichibu, T. Azuhata, T. Sota, and S. Nakamura, Appl. Phys. Lett. **69**, 4188 (1996); **70**, 2822 (1997).
- ⁴Y. Narukawa, Y. Kawakami, S. Fujita, and S. Nakamura, Phys. Rev. B 55, R1938 (1997).
- ⁵T. Takeuchi, S. Sota, M. Katsuragawa, M. Komori, H. Takeuchi,
- H. Amano, and I. Akasaki, Jpn. J. Appl. Phys., Part 2 36, L382 (1997).
- ⁶Y. Narukawa, Y. Kawakami, S. Fujita, and S. Nakamura, Phys. Rev. B **59**, 10283 (1999).
- ⁷K. P. O'Donnell, R. W. Martin, and P. G. Middleton, Phys. Rev. Lett. **82**, 237 (1999).
- ⁸A. Hori, D. Yasunaga, A. Satake, and K. Fujiwara, Appl. Phys. Lett. **79**, 3723 (2001); J. Appl. Phys. **93**, 3152 (2003).
- ⁹S. F. Chichibu, A. Uedono, T. Onuma, B. A. Haskell, A. Chakraborty,
- T. Koyama, P. T. Fini, S. Keller, S. P. Denbaars, J. S. Speck, U. K. Mishra, S. Nakamura, S. Yamaguchi, S. Kamiyama, H. Amano, I. Akasaki, J. Han, and T. Sota, Nat. Mater. **5**, 810 (2006).
- ¹⁰O. Brandt and K. H. Ploog, Nat. Mater. **5**, 769 (2006).
- ¹¹A. Hangleiter, F. Hitzel, C. Netzel, D. Fuhrmann, U. Rossow, G. Ade, and P. Hinze, Phys. Rev. Lett. **95**, 127402 (2005).
- ¹²H. Aizawa, K. Soejima, A. Hori, A. Satake, and K. Fujiwara, Phys. Status Solidi C 3, 589 (2006).
- ¹³U. Jahn, S. Dhar, M. Ramsteiner, and K. Fujiwara, Phys. Rev. B 69, 115323 (2004).
- ¹⁴Y. Takahashi, A. Satake, K. Fujiwara, J. K. Sheu, U. Jahn, H. Kostial, and H. T. Grahn, Physica E (Amsterdam) **21**, 876 (2004).
- ¹⁵N. Otsuji, K. Fujiwara, and J. K. Sheu, J. Appl. Phys. **100**, 113105 (2006).
- ¹⁶J. K. Sheu, G. C. Chi, and M. J. Jou, IEEE Photonics Technol. Lett. 13, 1164 (2001).