研　究　成　果　報　告　書

半導体量子構造系におけるキャリアー量子捕獲および離脱機構

課題番号　１６３６０１５７

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（基盤研究（B））　研究成果報告書

平成１９年６月

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この報告書は、平成１６年度から３年間にわたり実施したInGaN量子井戸構造を有する発光ダイオードの発光効率に関する研究をまとめたものである。エレクトロルミネッセンス効率の注入電流と温度依存性、フォトルミネッセンス発光効率の外部バイアスと温度依存性、光励起波長依存性、光励起強度依存性についての結果を総合的にまとめたものである。

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研究成果報告書の概要

InGaN系量子井戸発光ダイオードは、エピタキシャル成長層と基板間の格子不整合が大きいために10^{10}cm^{-2}程度の非常に多くの貫通転位が存在し欠陥密度が大きいにもかかわらず、発光効率が非常に高い。この高効率発光の機構を探るために、エレクトロルミネッセンス（EL）及びフォトルミネッセンス（PL）発光強度の外部電界効果を、（イ）EL発光効率の格子温度依存性及び（ロ）注入電流依存性、（ハ）PL発光効率の温度依存性及び（ニ）励起光波長依存性（直接励起と間接励起）、励起光強度依存性について、研究を実施した。その結果、以下の成果が得られた。

（１） エレクトロルミネッセンス（ＥＬ）発光効率の温度及び注入電流依存性の研究
青色及び緑色InGaN単一量子井戸発光ダイオードのEL発光強度の温度依存性(20-300K)から、特に、100K以下の低温で見られたEL強度の減少は、注入電流レベルが低いとき、また、In組成の異なる青色と緑色素子間で著しく変化する、即ち、注入キャリアーの捕獲効率の変化が重要であることを明らかにした。

（２） 電子溜層付加によるInGaN量子井戸ダイオードのEL発光効率と量子捕獲改善効果の研究
N型電子溜層の有無による青色多重量子井戸発光ダイオードのEL発光効率の温度依存性(20-300K)から、電子溜層の付加により、発光効率はすべての温度範囲、注入電流レベルで強められることを明らかにした。また、高注入電流と低注入電流時の比較から、主EL発光バンド効率の低温に於ける減少割合の変化及び短波長サテライトバンド発光効率の相関は、順バイアス電圧下でのキャリアー捕獲の違いにより起こ得ることを明らかにした。

（３） 緑色発光ダイドードのPL発光効率の外部電界効果とキャリアー捕獲及び離脱過程の研究
緑色InGaN単一量子井戸ダイオードのPL発光強度の外部電界効果を、直接励起及び間接励起条件下で測定した。直接励起条件では＋2V以上の順バイアス電圧で、また、逆バイアス条件下で、PL強度の減少が起こること、即ち、順バイアス電圧下でのキャリアー離脱による発光効率の減少が起こることを直接的に検証した。さらに、間接励起下では、＋2-3V程度の順バイアス下で発光効率が増大し、＋3.25V以上の順バイアス印加により著しく発光効率が減少する、即ち、障壁層から発光層へのキャリアー捕獲が発光効率に重要な役割を果たしていることを明らかにした。

（４） 緑色InGaN単一量子井戸発光ダイオードの発光効率と外部電界効果の研究
緑色InGaN単一量子井戸ダイオードのEL発光効率の100K以下の低温に見られた減少は、高注入電流時に顕著であるが、低注入電流時には起こらないこと、即ち、順バイアス電圧の大きさがキャリアーの発光中心への捕獲に重要な役割を演じていることを、直接励起下でのPL発光強度のバイアス電圧依存性との比較から明らかにした。

（５） 青色InGaN量子井戸発光ダイオードのPL発光効率と外部電界効果の研究
N型電子溜層付青色多重量子井戸発光ダイオードのPL発光効率外部電界効果の実験、特に、励起光強度依存性の実験から、電子溜層と活性層のPL強度比較により、逆バイアス下でのPL発光の減少は光生成した正孔の捕獲により律則されることを明らかにした。

CHAPTER 1

Impact of forward bias on the radiative recombination efficiency in blue and green InGaN quantum well diodes

CHAPTER 2

Electroluminescence efficiency of blue InGaN/GaN quantum well diodes with and without an n-InGaN electron reservoir layer

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Electroluminescence efficiency of blue InGaN/GaN quantum-well diodes with and without an *n*-InGaN electron reservoir layer

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The temperature dependence of the electroluminescence (EL) spectral intensity has been investigated in detail between $T=20$ and 300 K at various injection current levels for a set of two blue InGaN/GaN multiple-quantum-well (MQW) light-emitting diodes (LEDs) with and without an additional *n*-doped $In_{0.18}Ga_{0.82}N$ electron reservoir layer (ERL). The radiative recombination efficiency of the main blue emission band (~480 nm) is found to be significantly improved at all temperature regions and current levels when the additional ERL is introduced. For high injection currents I_f, i.e., large forward bias voltages V_f, a quenching of the EL intensity is observed for $T<100$ K for both LED structures, accompanying appearance of short-wavelength satellite emissions around 380–430 nm. Furthermore, the low-temperature intensity reduction of the main EL band is stronger for the LED without the ERL than with the ERL. For low I_f, i.e., small V_f, however, no quenching of the EL intensity is observed for both LEDs even below 100 K and the short-wavelength satellite emissions are significantly reduced. These results of the main blue emission and the short-wavelength satellite bands imply that the unusual evolution of the EL intensity with temperature and current is caused by variations of the actual potential field distribution due to both internal and external fields. They significantly influence the carrier capture efficiency by radiative recombination centers within the active MQW layer and the carrier escape out of the active regions into high-energy recombination centers responsible for the short-wavelength satellite emissions.
[DOI: 10.1063/1.2398690]

I. INTRODUCTION

Despite the great success of blue and green light-emitting diodes (LEDs) based on InGaN quantum-well (QW) heterostructures, the origin of the very bright emission characteristics is still controversially discussed.[1–10] A peculiar property of this material system is the observation of efficient luminescence at room temperature, although the density of misfit dislocations can be as high as 10^{10} cm^{-2} due to the large lattice mismatch between the InGaN/GaN epitaxial layers and the sapphire substrate. Therefore, we expect the existence of a particular mechanism, which is responsible for the enhancement of the radiative efficiency in the presence of a high defect density. Previous studies of the temperature-dependent electroluminescence (EL) spectral intensity in single-QW (SQW) diodes[7,8] show that efficient capture processes of injected carriers by localized tail states within the SQW layer play an important role for the recombination efficiency between $T=180$ and 300 K. However, for $T<100$ K, an anomalous quenching of the EL intensity is observed, which we attribute to a reduction of the vertical carrier capture rates.[7] We have also investigated the temperature dependence of the EL intensity for a specially designed blue InGaN/GaN multiple-QW (MQW) LED containing an additional *n*-doped InGaN electron reservoir layer (ERL).[11] This LED exhibits a significant improvement of the radiative efficiency in comparison with the LED without the ERL. In relation to the high luminescence efficiency under the presence of the high defect density in InGaN MQWs, Hangleiter *et al.*[12] and Hitzel *et al.*[13] recently proposed that most of the defects in the active region can be decorated by the higher band gap materials thus blocking the carriers to be nonradiatively extinguished, leading to the improved radiative recombination in spite of the existence of high defect density. This lateral carrier blocking mechanism against defect trapping within the active quantum-well layer is involved with the emissions at short-wavelength regions. Therefore, it is interesting to seek for whether the short-wavelength satellite emissions are correlated with the main EL band when the radiative recombination efficiency is significantly changed as a function of temperature and bias due to the changes of carrier capture efficiency by the radiative recombination centers.

In this paper, we report on a detailed investigation of the EL intensity of InGaN/GaN LEDs with and without the ERL as a function of temperature and injection current level to study how the carrier capture efficiency varies with bias conditions. By comparing the temperature-dependent EL charac-

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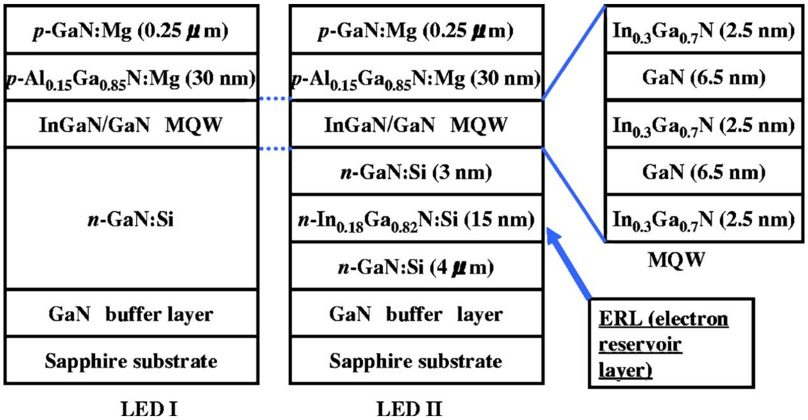

FIG. 1. (Color online) Schematic layered structure of the InGaN/GaN MQW-LEDs without (LED I) and with an additional n-type $In_{0.18}Ga_{0.82}N$ ERL (LED II).

teristics of the two MQW-LEDs for low and high injection currents over a wide spectral range, the enhanced EL efficiency observed in the MQW-LED with the ERL is attributed to an improved electron capture by radiative recombination centers. This capture is significantly influenced by the applied forward bias conditions (external field effects). Good correlation between the main EL efficiency and the short-wavelength satellite emission band is obtained.

II. EXPERIMENT

A set of two InGaN/GaN MQW-LEDs without and with an additional n-doped $In_{0.18}Ga_{0.82}N$ electron reservoir layer (ERL), named LED I and LED II, respectively, were grown by metal-organic vapor-phase epitaxy.[14] A schematic layered structure of the LEDs is shown in Fig. 1. The emission region of the LEDs 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. This MQW layer is clad by 4 μm n-GaN and 30 nm p-$Al_{0.15}Ga_{0.85}N$ layers. A 15-nm-thick n-doped ($\sim 10^{19}$ cm^{-3}) $In_{0.18}Ga_{0.82}N$ ERL is located between the n-GaN clad and the active MQW layer. A thickness of n-doped GaN barrier between the ERL and the active layer is thinned to be 3 nm. The p-i-n diode is formed by forming metal contacts on 0.25 μm p-GaN cap layer and the n-GaN clad layer laterally.[14] Thus, in this set of two diodes (LED I and II), only the addition of the ERL between the active MQW and the n-GaN barrier layers is different. EL spectra of the MQW-LEDs mounted on a Cu cold stage of a closed-cycle He cryostat were recorded for the dominating blue MQW emission band as well as for high-energy bands from other layers by conventional lock-in detection techniques at temperatures between 20 and 300 K as a function of the injected current between 1.0 and 50 mA.

III. RESULTS AND DISCUSSION

The current-voltage (I_f-V_f) characteristics of LEDs I and II have been measured between $T=20$ and 300 K. Figure 2 shows representative I_f-V_f curves at 20, 140, and 300 K. At 300 K, a typical V_f value for $I_f=10$ mA for LED II is 2.9 V, while for LED I the corresponding value of V_f is 3.6 V, so that a smaller V_f is necessary to achieve the same value of I_f for LED II with the ERL than for LED I without the ERL. That is, the forward bias voltage to get a certain current level is significantly reduced for LED II. This means that the standard Shockley recombination theory[15] needs to be modified by the additional heterostructure configuration. When the lattice temperature is decreased to 20 K, the I_f-V_f characteristics also change significantly, and the forward bias to obtain a current level increases substantially probably due to the reduced density of holes because of trapping by the deep Mg accepter level. That is, there are fewer holes in the p-type region at lower temperatures than at room temperature. At 20 K, the difference between the values of V_f for the two LEDs becomes even larger. When the temperature is decreased from 300 to 20 K in Fig. 2, the forward bias to achieve a sufficient current level, say 10 mA, is increased by about 2–3 V in both LEDs. This increase of the forward bias with decreasing temperature is most probably due to the reduced hole conductivity in the p-GaN and p-(Al, Ga)N layers at lower temperatures. In addition, we find a clear difference in the slope of the logarithm of I_f as a function of V_f between the two samples as well as between room and low temperatures (not shown here). Further investigations are necessary to fully understand the observed temperature dependence of the I_f-V_f characteristics and the different behaviors of the two LEDs. It should be noted, however, that the trapping of holes at 20 K due to the deep Mg accepter level ($\sim$170 meV) in p-GaN cannot explain all the observed

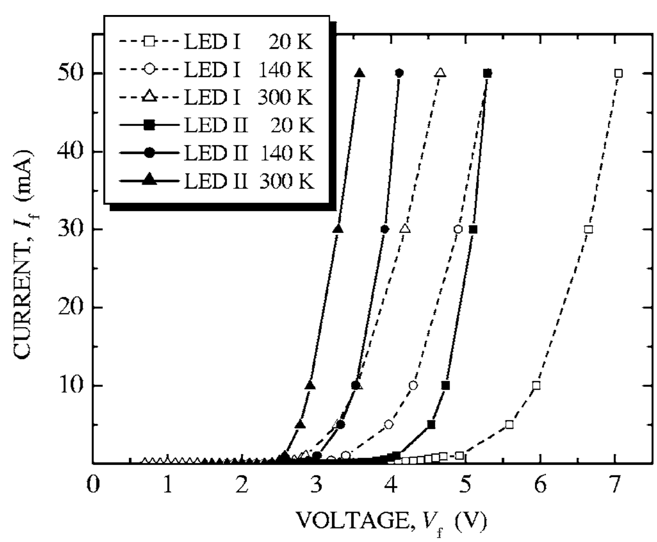

FIG. 2. I_f-V_f curves at 20, 140, and 300 K of the InGaN/GaN MQW-LEDs without (LED I: open symbols) and with an additional n-type $In_{0.18}Ga_{0.82}N$ ERL (LED II: full symbols).

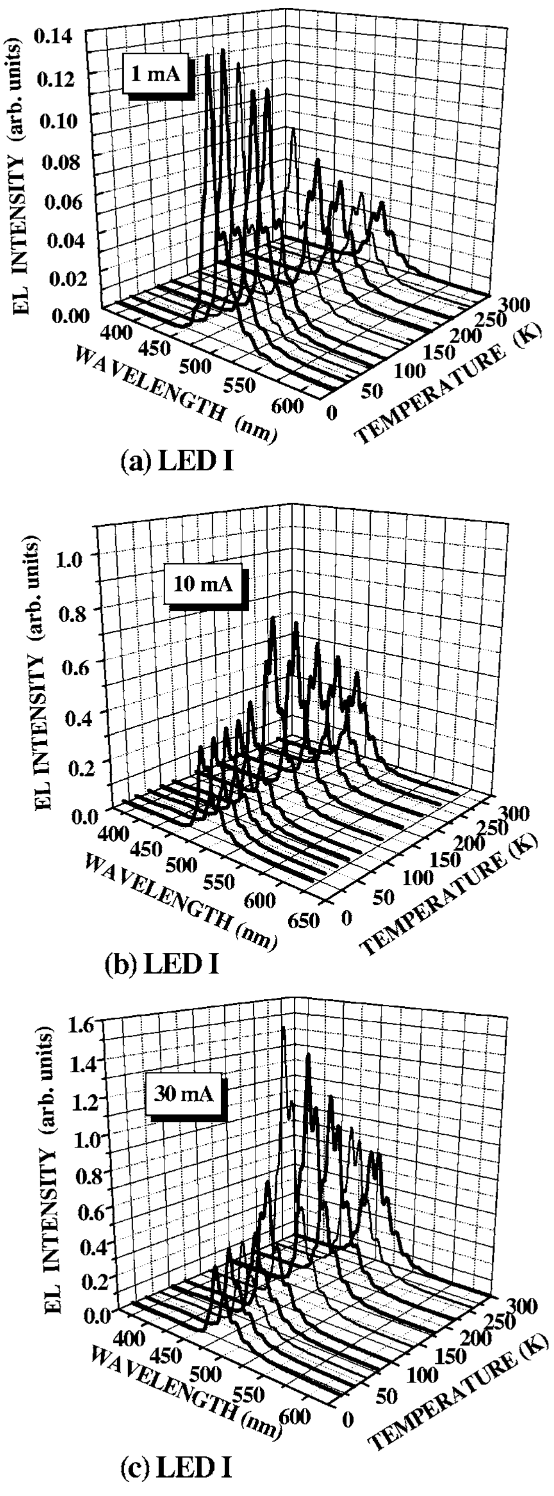

FIG. 3. Temperature-dependent EL spectra of LED I without an additional ERL for (a) I_f=1 mA, (b) 10 mA, and (c) 30 mA.

changes between LED I and LED II, since the *p*-GaN and *p*-(Al, Ga)N layers (with hole concentrations of $\sim 10^{17}$ cm^{-3}) are nominally identical in both samples. Here, we would like to emphasize that the addition of a wide *n*-$In_{0.18}Ga_{0.82}N$ layer between the *n*-GaN barrier and the MQW layer has a significant impact on the I_f-V_f characteristics, i.e., it reduces V_f for a particular value of I_f.

Figure 3 shows EL spectra of LED I without the ERL as a function of temperature at three injection current levels: (a) 1.0 mA, (b) 10 mA, and (c) 30 mA. The EL spectra exhibit intense emission around 480 nm with additional fine structures due to Fabry-Pe´rot interference fringes. When the injection current level is low at 1.0 mA in Fig. 3(a), the leading EL band exhibits the highest intensity at 20 K, and the EL peak intensity decreases as the temperature is increased. This decrease of the EL intensity with increasing the temperature is ascribed to an enhancement of nonradiative recombination processes, i.e., a reduction of the radiative recombination efficiency. When the current level is increased, however, the temperature dependence of the EL intensity is drastically changed, as illustrated in Figs. 3(b) and 3(c). A reduction of the EL intensity is clearly seen with decreasing the temperature below 100 K after reaching the maximum EL intensity around 140 K. At 30 mA [Fig. 3(c)] the EL intensity reduction is significant at 20 K and the difference in EL intensity between 140 and 20 K becomes larger than at 10 mA [Fig. 3(b)]. The similar temperature dependence of EL spectra is plotted in Fig. 4 for LED II with the ERL. At 1.0 mA [shown in Fig. 4(a)] the EL intensity observed as a function of temperature shows the similar behavior as the LED I, that is, the reduction of EL intensity with increasing the temperature. The maximum of the spectrally integrated EL intensity is reached at 20 K in Fig. 4(a). This enhancement of the radiative recombination efficiency around 20 K is commonly observed for both types of LEDs, which is similar to the photoluminescence (PL) efficiency enhancement observed at low temperatures due to the reduced nonradiative recombination.[5,10] However, it is important to note that the absolute EL intensity for LED II becomes larger than that for LED I for all temperatures (note the ordinate scale change). When the injection current level is increased to 10 mA, the difference in EL temperature dependence between LED I and LED II becomes even larger. That is, the EL reduction with decreasing temperature is strongly suppressed at 10 and 30 mA in Figs. 4(b) and 4(c) in comparison with Figs. 3(b) and 3(c). The LED II with the ERL always shows a stronger EL intensity than LED I without the ERL, in agreement with our previous results.[11,14]

In order to further investigate the origin of the reduced EL efficiency at lower temperatures and higher injection levels, we have also evaluated the spectrally integrated EL intensity divided by the current, that is, the EL external quantum efficiency as a function of temperature for different injection current levels. Figures 5(a) and 5(b) show the integrated EL intensity as a function of temperature at various current levels from 1.0 to 50 mA (normalized to the case of 10 mA) for LED I and LED II, respectively. For I_f =1.0 mA, the EL efficiency exhibits its highest value at 20 K, which monotonously decreases with increasing temperature in both cases. When the injection level is increased to 10 mA, the EL efficiency per injection current decreases in comparison with the case of I_f=1.0 mA. Note that the EL efficiency effectively decreases with increasing current level up to 50 mA for both LEDs. Therefore, we conclude that the reduction of V_f for LED II in comparison with LED I (cf. Fig. 2) plays an important role in the enhancement of the EL efficiency at 20 K. We emphasize that the EL efficiency per injected carrier is highest at the lowest current of 1.0 mA at

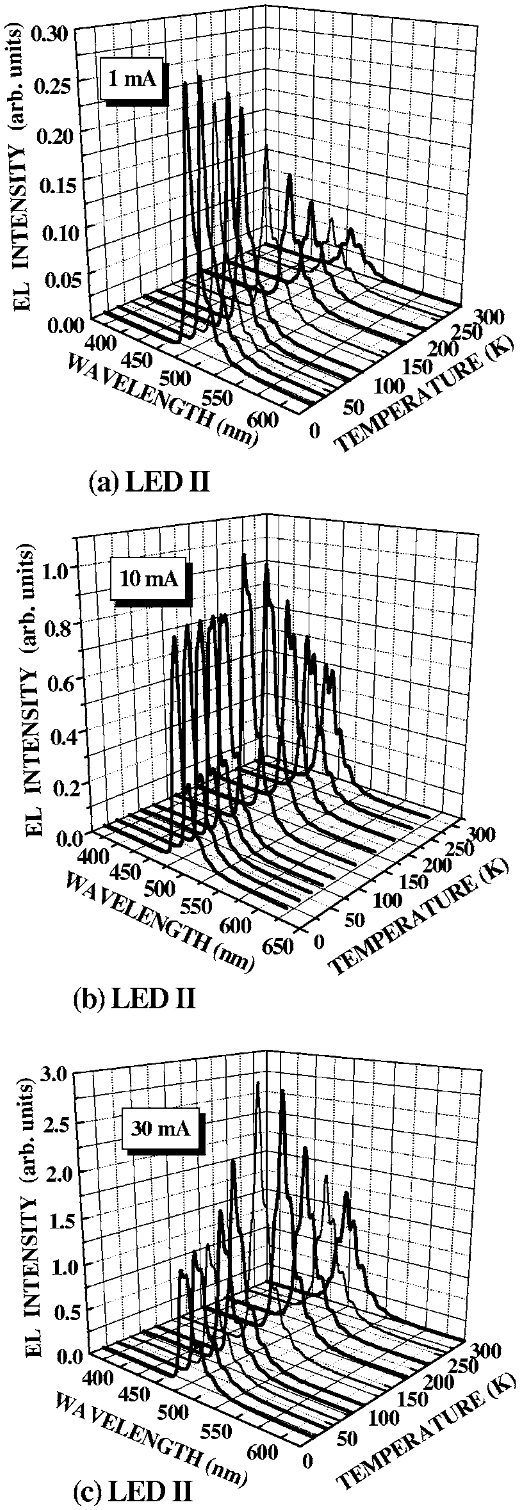

FIG. 4. Temperature-dependent EL spectra of LED II with an additional ERL for (a) I_f=1 mA, (b) 10 mA, and (c) 30 mA.

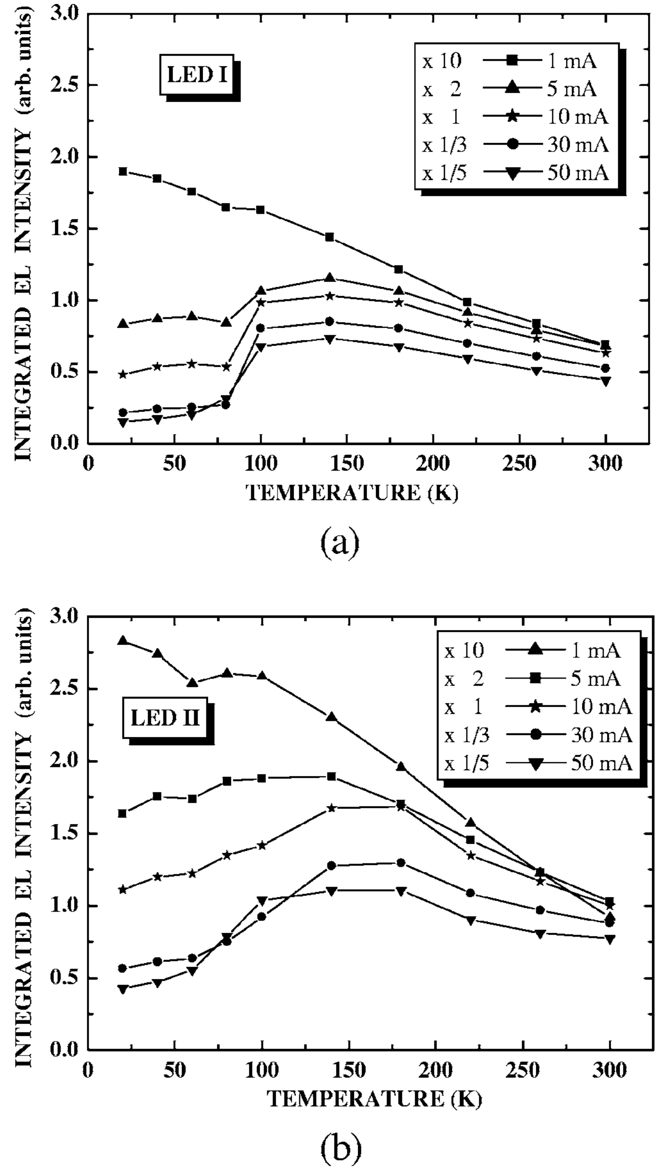

FIG. 5. Temperature dependence of integrated EL intensity of (a) LED I without and (b) LED II with an additional ERL for I_f=1, 10, 30, and 50 mA.

all temperatures in Fig. 5 for both of the diodes. It appears that the carriers are effectively captured by active centers in the MQW under the application of lower forward biases. But, applying higher forward biases, they are rather transferred to nonradiative recombination centers as a result of escape from the MQW region, thus reducing the EL efficiency. This is because the carriers can escape out of the well region due to the internal field effects, since the junction field direction is opposite to the internal field. We also note that the higher field existing in the well under the higher forward bias decreases the radiative recombination rate due to the quantum-confined Stark effect, which also causes the reduced EL intensity.

In Fig. 5, it is noted that the difference in the EL intensity between the two LEDs becomes even larger at 20 K for I_f=30 mA than for I_f=1.0 mA (cf. also Figs. 3 and 4). The difference of a factor of about 2 is seen between the EL intensities in Figs. 3(a) and 4(a) as well as in Figs. 3(c) and 4(c). For I_f=1.0 mA the ratio of the EL intensities of LED II to LED I is approximately temperature independent. But it strongly increases with decreasing temperature for I_f =30 mA. In order to demonstrate this difference more clearly, the ratio of the integrated EL intensity of LED II to LED I is plotted in Fig. 6 for I_f=1.0, 10, 30, and 50 mA as a function of temperature. Note that the EL intensity of LED II is always stronger than that of LED I, irrespective of temperature and current level. Furthermore, the enhancement of

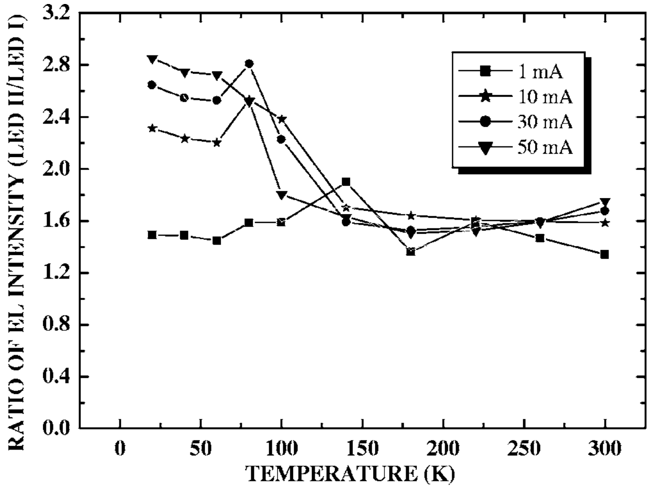

FIG. 6. Temperature dependence of ratio of integrated EL intensity (LED II/LED I) for $I_f=1$, 10, 30, and 50 mA.

the EL efficiency with decreasing temperature (around 20–80 K) is significant for LED II over LED I. The improvement of the EL efficiency for LED II with the ERL is, in fact, remarkable, indicating the importance of the carrier escape processes from radiative recombination centers in the InGaN MQW layer under the presence of higher forward biases.

In order to verify the carrier escape from the active MQW layer to the barrier regions, we have measured short-wavelength EL spectra around 360–450 nm below the leading blue (~480 nm) EL band including the spectral regions of the barrier materials and the ERL. The results are shown in Fig. 7 for LED I [(a) and (b)] and for LED II [(c) and (d)] at low (1.0 mA) and high (10 mA) currents, respectively. In Fig. 7(a) for LED I and at 1.0 mA, more than 100 times weaker emissions are seen at short-wavelength regions [peaked around 420 nm (2.95 eV)] than the main blue (~480 nm) band. The intensity of the satellite EL band monotonously increases with decreasing temperature, while the main EL band shows appreciable increases in intensity at 1.0 mA. Therefore, the results in Fig. 7(a) are explained by the fact that the EL efficiency is basically determined by the internal quantum efficiency due to the reduced nonradiative recombination processes. When the current is increased to 10 mA in Fig. 7(b), however, the EL intensity of satellite emissions drastically increases by almost one order of the magnitude relative to the main band and more rapidly increases when decreasing temperatures below 100 K. The EL efficiency of the main blue band thus decreases at 20 K when the current increases, as indicated in Fig. 5(a). That is, the main EL peak intensity decreases at temperatures below 140 K when the current is higher than 10 mA. On the other hand, as shown in Figs. 7(c) and 7(d), the short-wavelength satellite band of LED II is found to be much weaker than that of LED I. Enhancement of the satellite emission with increasing the current to 10 mA is not so significant in Fig. 7(d), indicating the improved carrier capture efficiency by the active region. This result is consistent with the improved radiative recombination efficiency in Figs. 4(b) and 4(c), especially at lower temperatures. But it is worth to note that the satellite EL emission also rapidly increases below 100 K in Fig. 7(d) in a similar way as in Fig. 7(b).

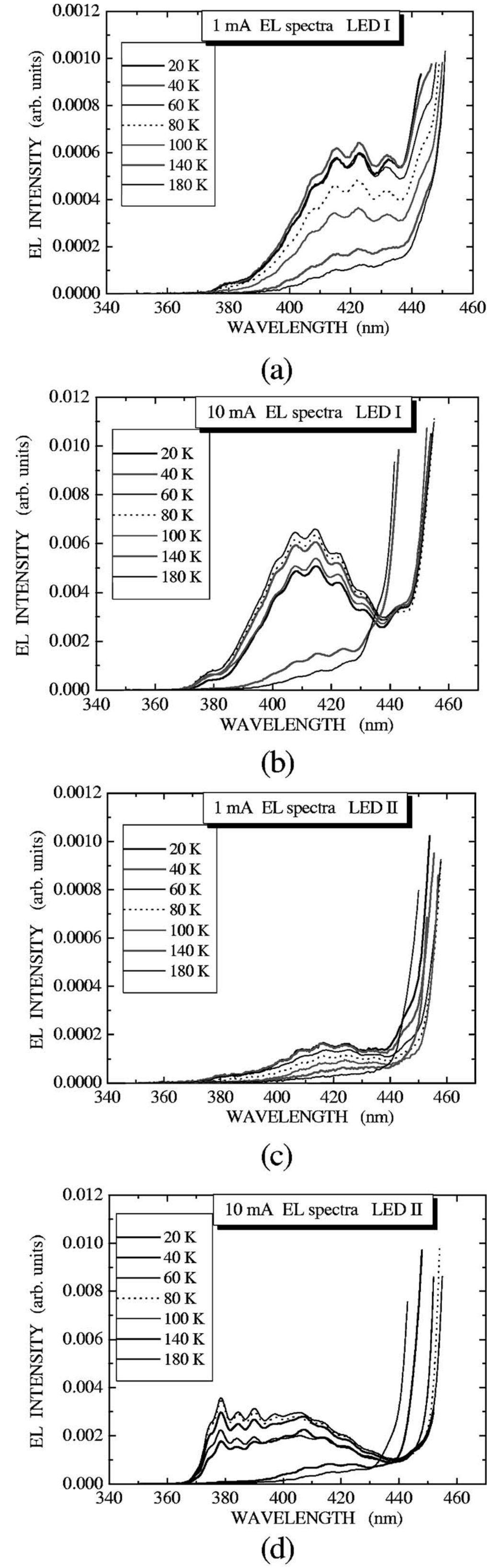

FIG. 7. Short-wavelength EL spectra of [(a) and (b)] LED I without and [(c) and (d)] LED II with an additional ERL for $I_f=1$ and 10 mA, respectively.

Figure 8 shows the satellite EL emission efficiency by

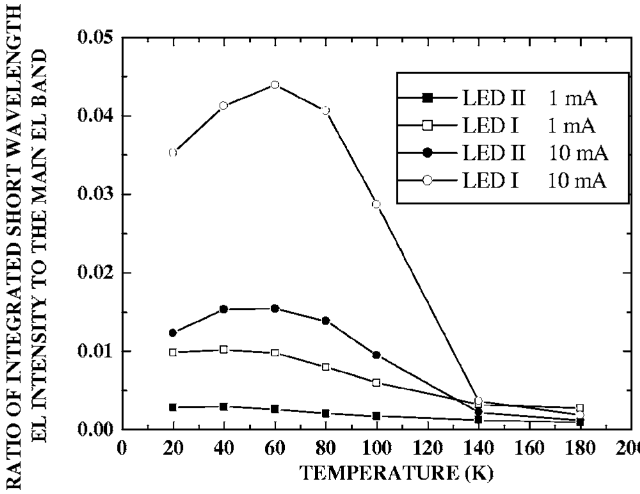

FIG. 8. Temperature dependence of ratio of the integrated short-wavelength EL intensity to the main EL band of LED I without and LED II with an additional ERL for $I_f=1$ and 10 mA.

plotting ratio of the integrated short-wavelength EL intensity divided by the integrated main EL intensity for LED I and II at low (1.0 mA) and high (10 mA) current levels as a function of temperature. For the injection current of 10 mA the short-wavelength emission efficiency of LED I is, in fact, increased at lower temperatures (below 100 K), indicating a reduction in the carrier capture rates by the MQW layer.[11] For LED II this increase of the satellite emission at 10 mA is significantly reduced and therefore the EL efficiency is improved due to the increased carrier capture efficiency. We note in Figs. 7 and 8 that, as the temperature goes down to 20 from 60 K, the short-wavelength satellite EL intensity slightly decreases. This is especially true for the injection current of 10 mA, while the main EL band is decreasing. We attribute these simultaneous intensity decreases of the main and satellite EL bands at 20 K to the reduction of the carrier injection efficiency from the clad layers into the active/barrier regions due to the decreased carrier mobility (most probably due to the decreased hole mobility).

In Fig. 7 the broad satellite EL band at short wavelengths around 370–430 nm (2.88–3.35 eV) is definitely enhanced under the higher forward bias conditions. It is important and interesting to inquire what the origin of this broad EL band is and whether the satellite band is related to the short-wavelength PL band (3.0–3.3 eV) observed by Hitzel *et al.*[13] for the high-band-gap material, effective as the carrier blocking layer against defect trapping. In our analysis the EL efficiency of the satellite band, which sensitively changes with bias and temperature, is simply used as a measure for the carrier escape out of the active regions in the MQW layer. Since our observation shows that the satellite EL band is significantly suppressed by the addition of ERL, electrons injected from the *n*-type ERL are more efficiently captured into the active regions under the forward bias conditions. This is because the band gap energy of the ERL material (estimated to be ~3.06 eV by a sharp PL peak observed) is lower than the GaN barriers (3.5 eV), so that the carrier overshoot is suppressed, thus enhancing the main blue EL band. One possibility for those overshooting electrons without the ERL is that they are to be captured in the *p*-GaN cap layer. Thus, the satellite emissions might be due to the donor-acceptor (D-A) pairs in the *p*-GaN cap layer. However, the broad (the bandwidth ~0.47 eV) satellite emission band and the large Stokes shifts (0.15–0.62 eV) observed are not consistent with the D-A recombination, as pointed out by Yang *et al.*[10] As a matter of fact, the satellite EL band is enhanced with increasing the forward bias and without the ERL. So, this emission band should not be related to the InGaN alloy fluctuations in the well layers (because the alloy effects are the same for both samples), but rather related to the barrier GaN layers. It is interesting to point out that the thinner sidewall quantum well as observed by transmission electron microscope[12] may also contribute to the stronger radiative recombination at the short-wavelength EL band. The observed broad linewidth for the satellite EL band may be explained if we take the quantum-confined Stark effect (QCSE) due to the internal field under the forward bias into account. However, more work is obviously necessary to elucidate the detailed carrier escape mechanisms under the forward bias conditions. Nevertheless, it is clear that the carrier capture by the MQW active regions is significantly improved by the additional ERL and by reducing the forward bias voltage, thus enhancing the EL efficiency by current injection.

IV. CONCLUSION

The EL spectral intensity of the main blue and the short-wavelength satellite bands has been investigated as a function of temperature and injection current for a set of two $In_{0.3}Ga_{0.7}N/GaN$ triple-quantum-well light-emitting diodes without and with an additional *n*-doped $In_{0.18}Ga_{0.82}N$ electron reservoir layer. We find that the temperature variation of the EL efficiency critically depends on the injection current level and the presence of the additional ERL. For low current levels and thus small forward bias voltages, EL quenching does not occur below 100 K due to efficient carrier capture, as evidenced by the reduced satellite EL band. However, for high injection current levels and thus large forward bias voltages, the EL quenching persists below 100 K, and its temperature variation is more pronounced in the LED without the ERL. This unique temperature dependence of the EL intensity variation at different injection levels originates from the difference in the forward bias voltage. Appearance of the short-wavelength satellite band when the main blue emission band is reduced is consistent with the EL mechanism by Hangleiter *et al.*[12] and Hitzed *et al.*[13] These results imply that the unusual evolution of the EL efficiency with current level and temperature can be caused by variations of the potential field distribution due to both internal and external fields, which significantly influence the carrier capture efficiency within the MQW active layer.

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CHAPTER 3

Carrier capture and escape processes in InGaN single quantum well diode under forward bias condition by photoluminescence spectroscopy

CHAPTER 4

Interplay of external and internal field effects on radiative recombination efficiency in InGaN quantum well diodes

CHAPTER 5

External field effects on photoluminescence properties of blue InGaN quantum well diodes

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

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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 excitation 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 $\sim 1-10^2$ W/cm^2).

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 MQW 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

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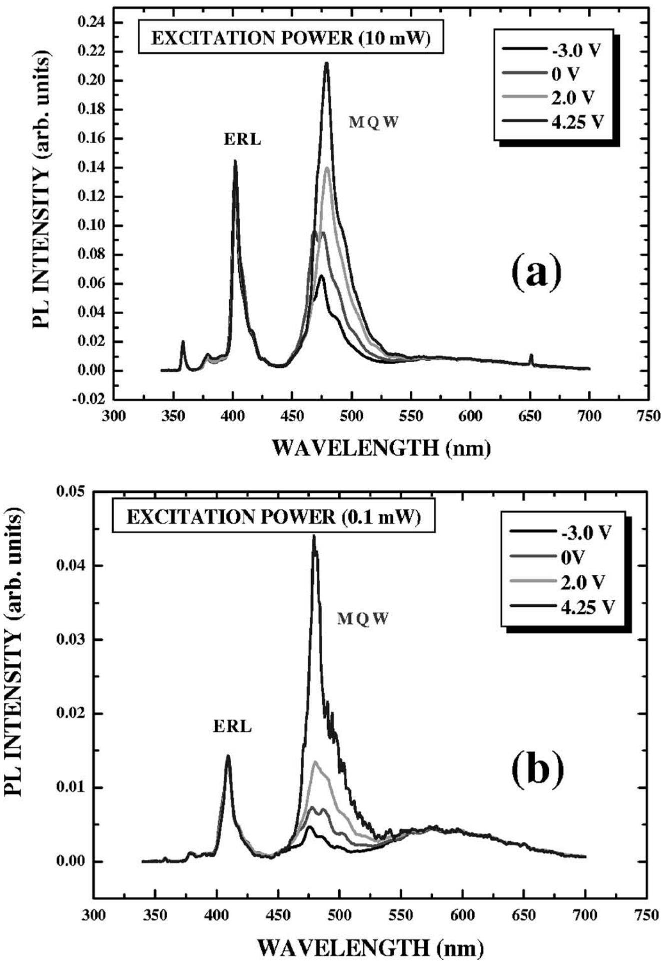

FIG. 1. PL spectra of a blue $In_{0.3}Ga_{0.7}N$ MQW-LED with an additional n-type $In_{0.18}Ga_{0.82}N$ electron reservoir layer (ERL) as a function of forward (positive) and reverse (negative) bias voltages at 20 K under (a) intense (10 mW power) and (b) weak (0.1 mW power) indirect excitations at 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.

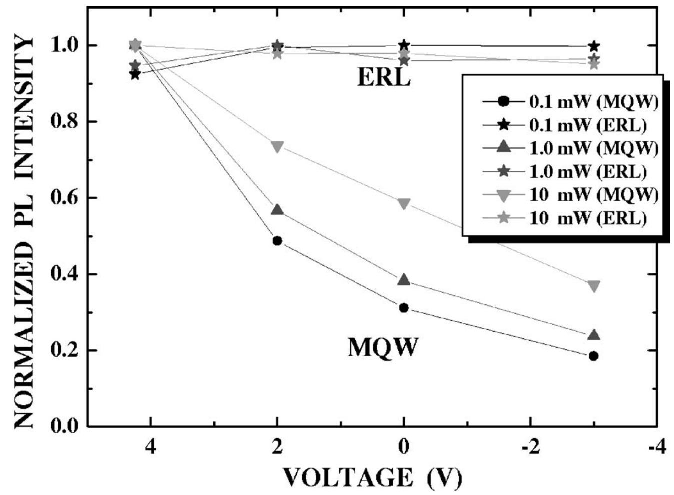

FIG. 2. Normalized, wavelength-integrated PL intensity for MQW and ERL bands as a function of reverse (negative) bias voltage at three different excitation powers of 0.1, 1.0, and 10 mW.

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 process 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 ($\sim 10^{19}$ 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

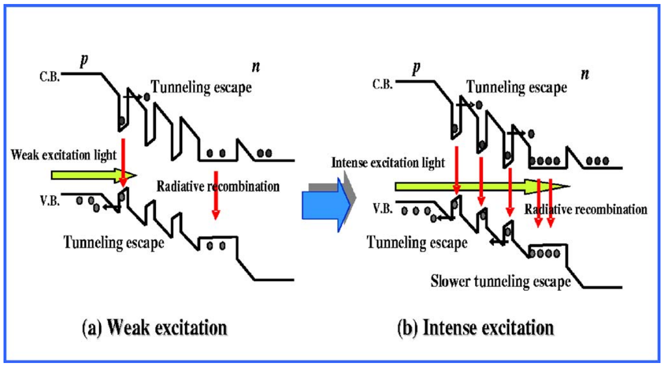

FIG. 3. (Color online) (a) Under the weak excitation the photon penetration depth is shallow, so that the photoexcited holes from the active MQW regions can escape more easily, leading to the rapid decreases of the PL intensity with increasing the reverse bias. (b) Under the intense excitation the MQW 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 InGaN 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.

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CHAPTER 6

Significance of vertical carrier capture for electroluminescence efficiency in InGaN multiple-quantum well diodes

CHAPTER 7

Abstracts for international conferences and domestic meetings