Optical Characteristics of InGaN Quantum Wells for High-Power and High-Efficiency True Green Laser Diodes

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Optical characteristics of InGaN quantum wells (QWs) for green laser diodes on semi-polar $\{20\bar{2}1\}$ GaN substrates were assessed using time-resolved photoluminescence (TRPL) and scanning near-field optical microscopy (SNOM). The InGaN QWs exhibited a remarkably shorter PL lifetime of 3.1 ns compared with that of conventional c-plane InGaN QWs, indicating that the piezoelectric fields are reduced greatly on $\{20\bar{2}1\}$ planes. Furthermore, the characteristic energy E_0 was estimated to be as small as 15.1 meV, which is less than one third of the reported value for c-plane InGaN QWs. Since E_0 represents the localization depth in InGaN QWs, this result proves the high homogeneity of the In composition on $\{20\bar{2}1\}$ planes. This conclusion was also verified by the spatially uniform distribution of the PL intensity and wavelength obtained by SNOM. These features are essential for highly efficient emissions in the green spectral region, consequently suggesting that the semi-polar $\{20\bar{2}1\}$ plane is suitable for fabricating green laser diodes.

Keywords: GaN, green laser, time-resolved photoluminescence, scanning near-field optical microscopy

1. Introduction

Laser light sources of the primary red-green-blue (RGB) colors are expected to improve the performance of display devices in terms of brightness, contrast and color gamut. Whereas red and blue semiconductor laser diodes (LDs) have been commercially available among the three colors, green semiconductor LDs have long been difficult to fabricate. In 2009, we successfully developed the world's first true green LDs based on InGaN material technologies(1)-(4). Subsequently, the LD performance of the output power and reliability has also been improving greatly(5),(6), as described in the previous article "High-Power and High-Efficiency True Green Laser Diodes." In these achievements, the utilization of a novel GaN substrate with a crystal plane of $\{20\bar{2}1\}$ played a key role. This new crystal plane enabled us to overcome the following two technical issues relating to InGaN quantum well (QW[*1]) active regions, namely, piezoelectric fields and In compositional fluctuations, which have hampered the development of green LDs on conventional c-planes. We have identified such features of the $\{20\bar{2}1\}$ plane mainly through the LD characteristics. In this study, in order to clarify the carrier recombination dynamics in which both the piezoelectric fields and the In compositional fluctuations are closely involved, the optical properties of the $\{20\bar{2}1\}$ InGaN QWs were assessed using time-resolved photoluminescence (TRPL) and scanning near-field optical microscopy (SNOM), in collaboration with Kyoto University. On the basis of the insights obtained regarding the optical emission mechanisms, the advantages of the $\{20\bar{2}1\}$ plane and their correlation with the LD performance were discussed.

2. Technical Issues of InGaN QWs for Green LDs

Active layers of epitaxial structures for green LDs consist of ternary InGaN, which is a mixture of GaN and InN. To obtain green emissions in the spectral range of 525 to 535 nm, the In composition needs to be raised to about 30%, which brings about two technical issues owing to the large lattice mismatch between GaN and InN. One is the increase of the piezoelectric fields within InGaN QWs. When compressive in-plane strain is applied to InGaN QWs, positive charges of group III atoms (Ga and In) and negative charges of N atoms become no longer balanced, resulting in the generation of piezoelectric fields along the c-axis direction. This effect causes the band bending in the active region as shown in **Fig. 1 (a)**, and then diminishes

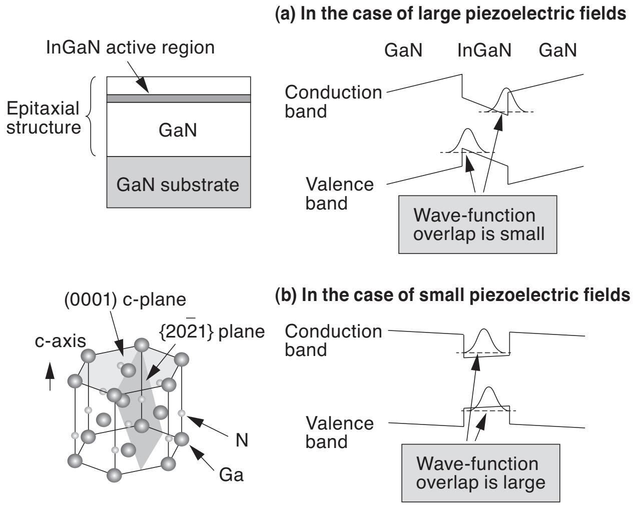

Fig. 1. Influences of piezoelectric fields in InGaN QWs

the electron and hole wave-function overlap, leading to the lowering of the radiative recombination probability(7),(8).

The other issue is the enhancement of the In compositional fluctuations. Since GaN and InN exhibit immiscibility owing to the above-mentioned large lattice mismatch(9), the In compositions spontaneously become inhomogeneous. In such a case, local potential minima, which correspond to locally In-rich regions, are created in the energy levels of InGaN QWs as shown in **Fig. 2**. Parts of injected carriers (electrons and holes) are diffused to the lower-lying energy levels of these localized states. As for blue light-emitting diodes (LEDs*2) with relatively low In contents (about 15%), this carrier localization phenomenon contributes to the improvement of the emission efficiency because they prevent carriers from being trapped by nonradiative centers(10),(11). However, when the In compositions are increased for green emissions, the In-rich regions were reported to trigger the defect formation accompanied by the thermally induced In segregation [**Fig. 2 (i)**](12). Thus, the carrier transfer to the localization states conversely deteriorates the emission efficiency for green LEDs. Furthermore, in the case of LDs, the In compositional fluctuations were shown to degrade the threshold current and slope efficiency because they result in the broadening of the photon energy levels and cause the adverse effect in light amplification [**Fig. 2 (ii)**](13). Therefore, the reduction of the In compositional fluctuations is essential for high In-content InGaN-based green LDs.

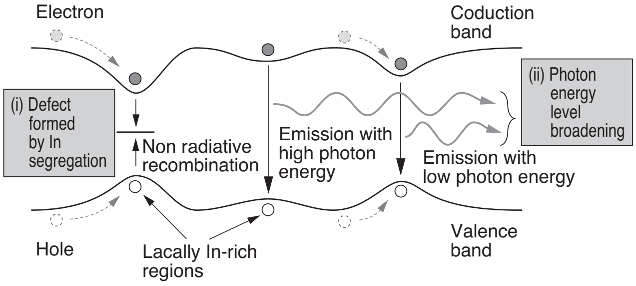

Fig. 2. Influences of In compositional fluctuations in InGaN QWs

Since the piezoelectric polarization is induced along the c-axis direction, its influence can be alleviated by fabricating laser structures on crystal planes inclined from the c-plane as shown in **Fig. 1 (b)**. Indeed, various non-polar and semi-polar planes, such as $\{10\bar{1}0\}$ (m-plane), $\{11\bar{2}0\}$ (a-plane) and $\{11\bar{2}2\}$, have been extensively studied, and their effectiveness has been demonstrated. However, the In compositional fluctuations have still remained an issue on these planes. On the other hand, laser structures grown on a semi-polar $\{20\bar{2}1\}$ plane exhibited the uniform microscopic photoluminescence (PL) image and the narrow luminescence line width, indicating that this plane exerts a favorable effect on the decreases not only in the piezoelectric fields but also in the In compositional fluctuations(1),(3). In this work, in order to clarify the emission mechanisms behind such characteristics of the $\{20\bar{2}1\}$ plane, the localized states were evaluated by TRPL and SNOM-PL.

3. Experimental

3-1 Sample structure

A 525-nm band single QW (SQW) structure was grown on an n-type $\{20\bar{2}1\}$ plane GaN substrate by organometallic vapor phase epitaxy (OMVPE*3). Free-standing $\{20\bar{2}1\}$ plane GaN substrates were produced by hydride vapor phase epitaxy (HVPE*4)(14). The threading dislocation densities were less than 1×10^6 cm^{-2}. A 2-μm-thick n-type GaN buffer layer was directly grown on the GaN substrate, followed by a 100-nm-thick undoped InGaN buffer layer, a 15-nm-thick undoped GaN barrier layer, a 3-nm-thick InGaN SQW, and a 10-nm-thick GaN capping layer.

3-2 Measurement procedures of TRPL and SNOM-PL

TRPL is a method of monitoring PL transients under pulsed excitation, and it provides lifetimes of excited states. The excitation pulse light source used here was a frequency doubled Ti:Al_2O_3 laser with a pulse repetition rate of 4 MHz and width of 1.5 ps. To selectively excite the InGaN QW, the excitation wavelength was tuned to 400 nm. The excitation power density was adjusted between 8.6 and 29,000 nJ/cm^2, while measurement temperature was changed from 6 K to room temperature (RT). The PL was detected with a streak camera.

Figure 3 (a) shows a model of localized states. The inhomogeneity in InGaN QWs causes the potential fluctuations as schematically shown in this figure. In such a case, the lifetime of PL with high photon energy becomes shorter than that with low photon energy because not only the radiative recombination process [**Fig. 3 (a) (i)**], but also the carrier transfer process to the tail states [**Fig. 3 (a) (ii)**], are involved in the decay of PL with low photon energy. The relation between the PL lifetime τ_{PL} and the emission photon energy E can be expressed by the equation(15):

$$\tau_{PL}(E) = \frac{\tau_{rad}}{1 + \exp\left(\frac{E - E_{me}}{E_0}\right)} ,$$

where E_0 is the characteristic energy representing the localization depth and E_{me} is the mobility edge, as shown in **Fig. 3 (b)**. τ_{rad} is the radiative recombination lifetime. Fitting the energy dependence of the PL lifetime with this model estimates E_0, which serves as a reference of the homogeneity of InGaN QWs. Smaller E_0 is beneficial for green LDs because it represents homogeneous InGaN QWs with smaller compositional fluctuations.

While this TRPL assesses macroscopic nature for carrier localization, SNOM reveals nano- or micro-scaled optical emission dynamics, as explained below. SNOM-PL is a microscopy technique, which provides PL imaging with high spatial resolution beyond the diffraction limit by utilizing evanescent light as a probe. Evanescent light is obtained by the optical interaction of light emerging from a sub-wavelength aperture, and thus the resolution is limited by the aperture size, not by the wavelength of the light source(16). In this study, a double-taper fiber probe

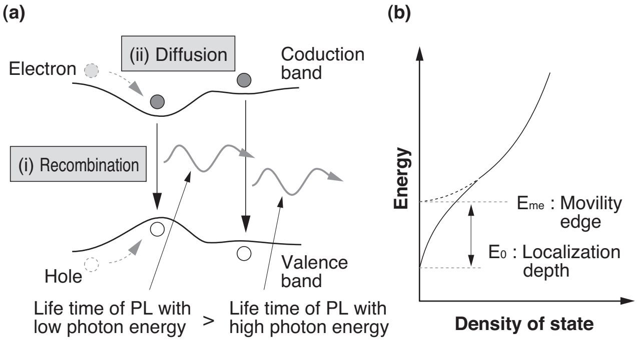

Fig. 3. Model of localization states

with an aperture diameter of 150 nm was employed for photo-excitation and PL probing, whereby the same order of the spatial resolution can be expected to be achieved. A cw InGaN LD emitting at 405 nm was used to selectively excite the InGaN QW. The excitation power density was about 7 kW/cm^2, which corresponds to a photo-generated carrier density of about 1×10^{18} cm^{-3}. The measurements were performed at RT.

4. Results and Discussion

4-1 Characterization of localization states by TRPL

Figure 4 shows the PL decay curve of the $\{20\bar{2}1\}$ InGaN SQW at 6 K. The excitation energy density was 2.6 μJ/cm^2, which corresponds to a photo-generated carrier density of 5×10^{16} cm^{-3}. This condition barely affects the internal electric fields because of the sufficiently low photo-generated carrier density$^{(17)}$. It was found that the PL decay curve on the $\{20\bar{2}1\}$ plane cannot be expressed by a single exponential function, which is similar to c-plane InGaN QWs. Assuming that the curve was composed of fast and slow decays, the intensity-dominant fast decay time was estimated to be 3.1 ns, which corresponds to a radiative recombination lifetime because nonradiative processes are negligible at 6 K. The radiative lifetime of 3.1 ns we achieved is much shorter than that of green InGaN QWs on the c-plane (85 ns)$^{(18)}$. The radiative lifetime is inversely proportional to the square of the electron and hole wave-function overlap. Therefore, this result is an indication that the wave-function overlap on the $\{20\bar{2}1\}$ plane is larger than that on the c-plane, proving that the piezoelectric fields are well reduced on the $\{20\bar{2}1\}$ plane.

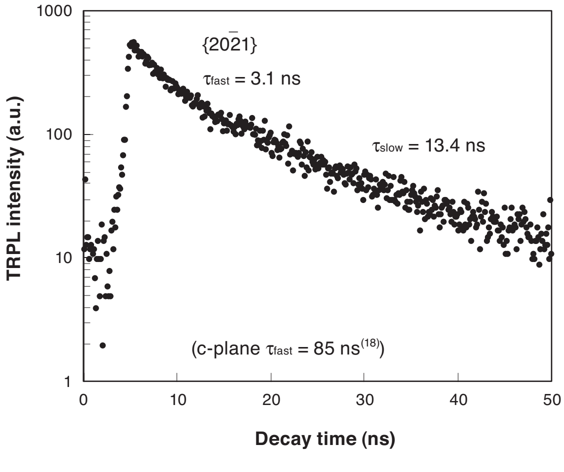

Fig. 4. PL intensity decay curve at 6 K

Next, the potential fluctuations on the $\{20\bar{2}1\}$ plane were evaluated by analyzing the PL lifetime with respect to the emission photon energy. **Figure 5** shows the energy dependence of the PL lifetime at **(a)** 6 K and **(b)** 300 K. The time-integrated PL spectra were also shown in the same figures. The PL lifetime at 6 K decreased for photon energies larger than the PL peak energy, which is a characteristic behavior of a localized electronic system as described in the previous section. Thus, this result suggests that localization states exist to some extent in the $\{20\bar{2}1\}$ InGaN SQW. Fitting the experimental data using the above-mentioned formula yields the characteristic energy E_0 as 15.1 meV. This value is about one third that for green InGaN QWs on the c-plane (51 meV)$^{(19)}$ and about half that for blue-violet InGaN QWs on the c-plane (33.3 meV)$^{(20)}$, indicating

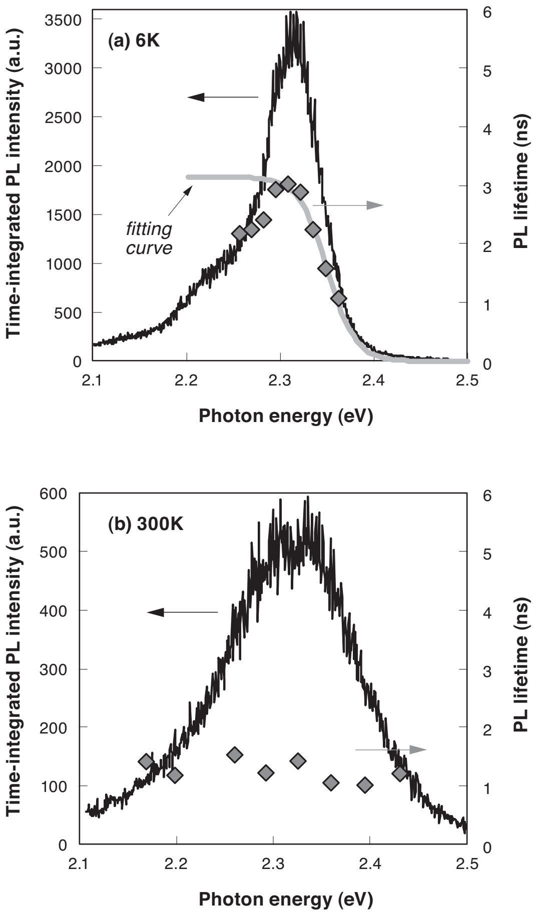

Fig. 5. PL spectra and energy dependence of the PL lifetime at (a) 6 K and (b) 300 K

that the localization depth on the $\{20\bar{2}1\}$ plane is remarkably shallow. Furthermore, the emission energy dependence of the PL lifetime began to decrease around 150 K, which corresponds to 13 meV, and fully disappeared at 300 K, as shown in **Fig. 5 (b)**. This finding indicates that a thermal energy greater than 150 K (13 meV) delocalizes carriers from the shallow localization states with E_0 = 15.1 meV. The estimated small localization energy and the consequent delocalization at elevated temperatures on the $\{20\bar{2}1\}$ plane are consistent with the homogeneous microscopic PL image of the LD structure at RT(1),(3). These results demonstrate that the utilization of the $\{20\bar{2}1\}$ plane leads to a marked reduction in the potential fluctuations in InGaN QWs.

In order to address this feature in more detail, the PL peak shift at 6 K with respect to the excitation power density was investigated, as shown in **Fig. 6**. The excitation power densities were changed between 8.6 and 29,000 nJ/cm^2, which correspond to photo-generated carrier densities from 1.7×10^{14} to 5.7×10^{17} cm^{-3}. The PL peak of the $\{20\bar{2}1\}$ InGaN SQW remained almost unchanged, while that of c-plane InGaN QWs shows a large blueshift greater than 60 meV(19). The PL peak is mainly affected by 1) the screening of the internal electric fields and 2) the filling of the localization states. Since the former cause is negligible in the present case owing to the sufficiently low photo-generated carrier densities(17), the latter factor is believed to be responsible for the observed distinct difference in the PL peak shift. The very small energy shift on the $\{20\bar{2}1\}$ plane can be interpreted as resulting from a considerably small density of band-tail states of homogeneous InGaN QWs, which supports the conclusion that we obtained through TRPL.

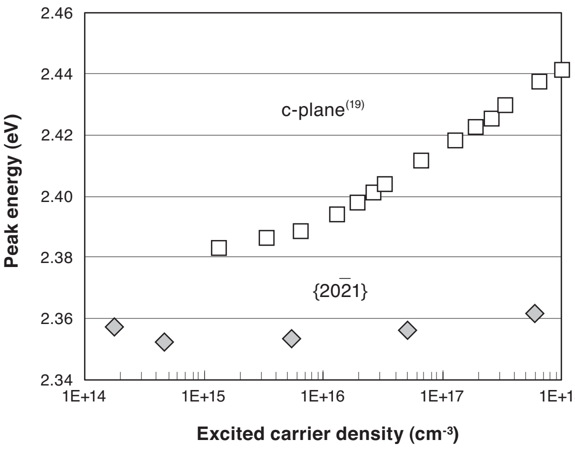

Fig. 6. Excited carrier density dependence of the PL peak energy

4-2 Characterization of local PL properties by SNOM

Now that the macroscopic optical properties have been revealed by TRPL, we next focus our attention on the local PL characteristics, which are also important for understanding the emission mechanisms of InGaN QWs as discussed in **Fig. 2**. The same $\{20\bar{2}1\}$ InGaN SQW was characterized by SNOM-PL in order to study nanoscopic optical properties. **Figures 7 (a) and (b)** show the spatial distribution of the integrated intensity and peak wavelength, respectively. The integrated PL intensity [**Fig. 7 (a)**] exhibited islandlike structures with diameters of a few hundred nanometers, which were almost comparable to the aperture diameter (150 nm) of the SNOM fiber probe. The ratio between the maximum and minimum intensity was less than 4. This is much smaller than the reported value of about 200 for a c-plane InGaN SQW(21). Furthermore, the spatial variation of the PL peak wavelength was about 5 nm [**Fig. 7 (b)**], which also gives an indication of homogeneous InGaN QWs on the $\{20\bar{2}1\}$ plane, considering that the c-plane SQW is reported to show the spatial variation in the PL peak wavelength of larger than 15 nm(21). Concerning stripelike distribution along the $[\bar{1}2\bar{1}0]$ direction in the mapping of the PL peak wavelength, it was found to correspond to the surface morphology by superimposing the SNOM-PL image with the atomic force microscopy image(22). These results clearly demonstrate that the $\{20\bar{2}1\}$ SQW has a spatially more uniform potential distribution than the c-plane SQW, which is consistent with the above-discussed macroscopic PL features. Such InGaN QWs with high homogeneity are advantageous in suppressing the defect generation originating from the In segregation as well as in reducing the photon energy level broadening, thus making the $\{20\bar{2}1\}$ plane beneficial for green LDs in terms of improving the threshold current and the slope efficiency.

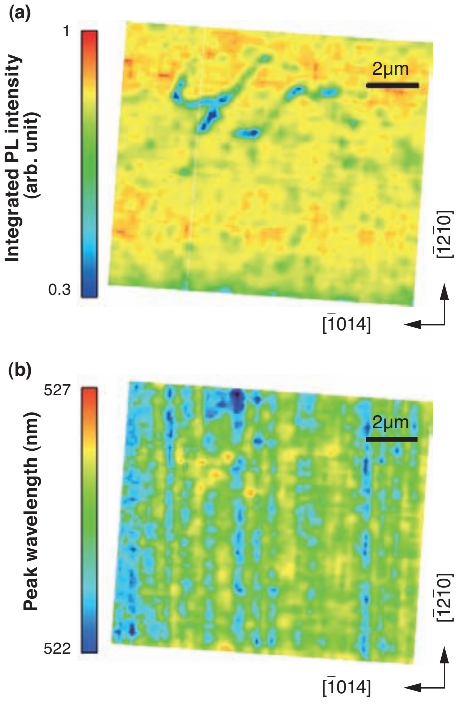

Fig. 7. Spatial distribution of (a) integrated intensity and (b) peak wavelength obtained by SNOM-PL

5. Conclusion

The optical characteristics of InGaN QWs on $\{20\bar{2}1\}$ planes were investigated by means of TRPL and SNOM-PL. TRPL revealed that the PL lifetime of $\{20\bar{2}1\}$ InGaN QWs is much shorter than that of conventional c-plane InGaN QWs, indicating that the piezoelectric fields are reduced greatly on $\{20\bar{2}1\}$ planes. In addition, the shallow localization depth derived from the energy dependent PL lifetime along with the small blueshift under weak photo-excitation proved that $\{20\bar{2}1\}$ InGaN QWs possess remarkably smaller compositional fluctuations compared with those on c-planes. This high homogeneity on $\{20\bar{2}1\}$ planes was also verified by the spatially uniform distribution of PL intensity and wavelength assessed by SNOM. Such high quality InGaN QWs obtained by exploiting these features have played a key role in the development of the world's first green LDs, as well as the subsequent improvements of the output power and the reliability. These experimental observations pursuing basic optical properties suggest that the $\{20\bar{2}1\}$ plane offers distinct advantages over the c-plane and is consequently suitable for fabricating green LDs. Based on the fundamental emission mechanisms elucidated in this work, the device performance of green LDs is expected to keep advancing toward further expansion of their application areas.

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Technical Terms

*1 QW (quantum well): A structure composed of a thin well layer with a nanometer-scale thickness sandwiched by barrier layers with larger band gap energy.
*2 LED (light emitting diode): A semiconductor device which converts electrical energy into light by current injection.
*3 OMVPE (organometallic vapor phase epitaxy): A chemical vapor deposition method of growing semiconductor crystals using organometallic precursors.
*4 HVPE (hydride vapor phase epitaxy): A chemical vapor deposition method of growing semiconductor crystals using hydride gases for group V precursors.

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