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Short Note

Spatial Inhomogeneity of Photoluminescence in an InGaN-Based Light-Emitting Diode Structure Probed by Near-Field optical Microscopy Under Illumination-Collection Mode

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Spatial distribution of photoluminescence (PL) spectra has been assessed in an InGaN single quantum well (SQW)-based light-emitting diode structure by near-field optical microscopy under the illumination-collection mode. The obtained PL mapping image revealed a variation in both peak and intensity of PL spectra according to the probing location with a scale less than about 200 nm. The variation in PL intensity is from 0.8 to 1.8 in arbitrary units indicating that the internal quantum efficiency fluctuates from 10% to 50% within the active layer.

KEYWORDS: InGaN, SNOM, illumination-collection mode, PL mapping image, spatial inhomogeneity

Recent progress in growth technology has led to the commercialization of InGaN-based light-emitting diodes (LEDs) in near ultraviolet (UV), violet, blue, green and amber spectral regions.[1–3] In spite of the high threading dislocation density (10^8–10^{10} cm^{-2}) in epilayers grown on sapphire, such LEDs exhibit a substantially high external quantum efficiency (η_{ext}). However, further improvement of η_{ext} is desired in order to extend the application area of LEDs. Mukai *et al.* reported the variation of η_{ext} driven under a forward current of 20 mA as a function of emission wavelength of InGaN-based LEDs with different In mole fractions.[4] Although the GaN LEDs emitting at 360 nm exhibited the η_{ext} of approximately 1%, the addition of In to active layers resulted in remarkable increase in the η_{ext} value, reaching about 11% at an emission wavelength of 460 nm. These phenomena have been understood as two major mechanisms. The first one is that nonradiative recombination centers (NRC), whose origins are probably not macroscopic defects but point defects, are suppressed by the substitution of In atoms to Ga-sites. The second one is the so-called localization effect, where excitons and/or carriers are trapped at deep energy states formed by large alloy fluctuations, so that the pathway to the NRC is hindered very effectively.[5–7] Nevertheless, it was also reported that η_{ext} of InGaN-based LEDs decreases if the emission wavelength increases longer than the blue-green region. Although this mechanism has not been clarified yet, microscopic or nanoscopic optical assessments would provide valuable information. Several reports have recently been published on spatial mapping measurements of emissions in InGaN single quantum wells (SQWs) by employing cathodoluminescence (CL)[8] and scanning near-field optical microscopy (SNOM).[9–13] It has been found that the lateral size of In-composition fluctuations is about 100 nm, which may be limited by the diffusion length of carriers, and/or by the resolution of spectroscopy. Most SNOM results were obtained using a laboratory-built illumination-mode system, where the laser light was focused on about 200 nm^2 spot through the SNOM tip composed of a tapered optical fiber and the photoluminescence (PL) spectra were probed using the objective lens. The disadvantage of this system is that it is very difficult to determine whether excitons and/or carriers producing PL are directly photogenerated in the probe region or diffused from outside. This disadvantage can be overcome by means of the illumination-collection mode, where photoexcitation and PL probing are performed using the same fiber tip. However, there have been no reports on this mode for the assessment of InGaN-based semiconductors because the Ge-doped SiO_2 core normally used for the SNOM tip exhibits broad PL by wave-guiding blue to near-UV laser beams. We report the first PL imaging of an InGaN-based green LED structure using SNOM under the illumination-collection mode. This was achieved by tailoring the tapered structure of the fiber tip composed of a pure SiO_2 core.

The sample used in this study was grown on a (0001)-oriented sapphire (Al_2O_3) substrate by a two-flow metalorganic chemical vapor deposition (MOCVD) technique.[14] The layers are a GaN buffer layer (30 nm), an n-GaN:Si layer (4 mm), an $In_{0.3}Ga_{0.7}N$ active layer (3 nm), a p-$Al_{0.2}Ga_{0.8}N$:Mg layer (100 nm) and a p-GaN:Mg layer (0.5 mm). The near-field PL measurements were performed using a NFS-300 near-field spectrometer with a He–Cd laser developed by JASCO Corp. The system consists of a probe, a feedback loop, and a monochromator. The probe was used to illuminate the sample by the excitation light and to collect PL from the sample. A near-field probe was fabricated by etching using a hydrofluoric-buffered solution, and an aperture with the diameter of 500 nm was formed on the 150-nm-thick Au coat. The fiber for this probe has the pure SiO_2 core for transmitting near-UV light with low transmission loss, and the emission background from the fiber is prevented effectively. Sample-probe separation was regulated by detecting the amplitude of the dithered probe. The amplitude of this oscillation was less than 1 nm at the first-order resonance frequency of the probe. This amplitude was fed back to control the height of the sample PZT stage. As a result, the sample-probe separation was regulated to be 10 nm. Although the UV-emission line from the He-Cd laser (325 nm) can be transmitted to the fiber, the blue line (441.6 nm) was used as the excitation source in order to achieve selective photoexcitation to an $In_{0.3}Ga_{0.7}N$ SQW. The optical power of 6 mW was coupled to the probe, and about 6 μW was used to illuminate the sample through the probe. PL collected using the probe (illumination-collection) was introduced into the single monochromator. The PL signal was detected by using a liquid nitrogen-cooled charge-coupled device detector. All measurements in this study were performed at room temperature.

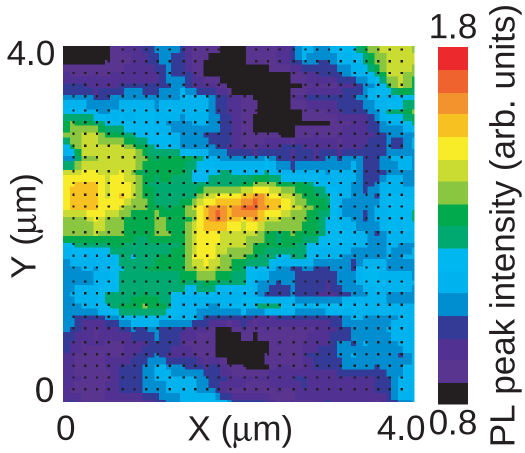

Fig. 1. Near-field PL image mapped with the PL peak intensity. Each dot plotted periodically at an interval of 0.13 μm shows the position of probing.

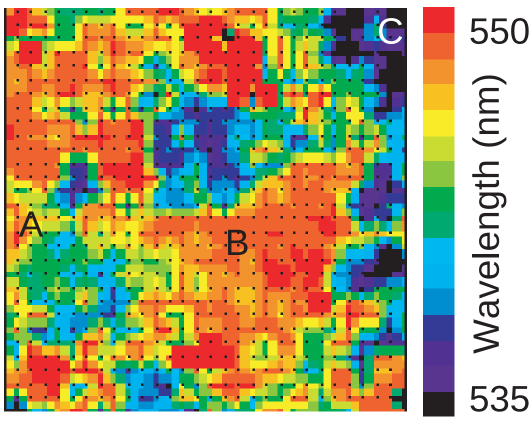

Fig. 2. Near-field PL image mapped with the PL peak wavelength.

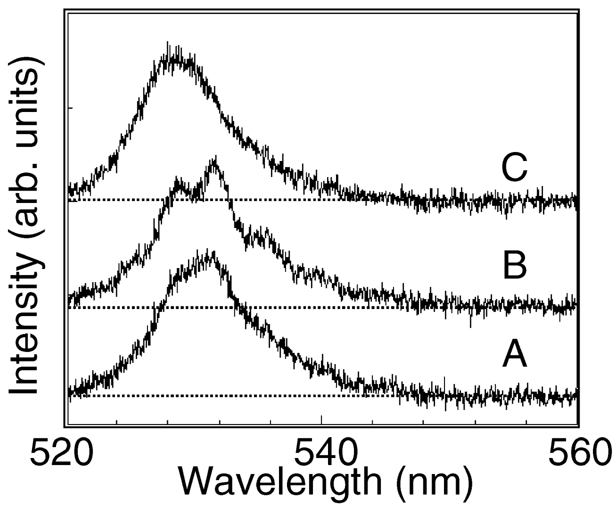

Fig. 3. PL spectra taken at positions A, B and C, which are marked in Fig. 2.

Near-field PL images of peak intensities were taken at an InGaN SQW LED structure by scanning a 4 μm × 4 μm area with the mapping interval of 0.13 μm. The aperture of the probe is about 0.5 μm in diameter. The InGaN SQW active layer is located about 0.6 μm beneath the sample surface. However, spatial inhomogeneity with the size of the mapping interval is observed as shown in Fig. 1. Similar results have also been observed in other semiconductors.[15] It is assumed that the close proximity of the glass apex to the sample, the large cone angle of the tip, and the modal occupation of the tapered region contribute to the realization of a high numerical aperture lens.[15] Figure 2 shows the image mapped with the PL peak wavelength. It was observed that the PL peak wavelength fluctuated by about 15 nm probably due to the inhomogeneous distribution of In the alloy composition. The PL spectra at positions A, B and C marked in Fig. 2 are depicted in Fig. 3. The integrated PL intensity fluctuated from 0.8 to 1.8 in arbitrary units. There was no clear spatial correlation between PL intensity and peak wavelength. Simple calculation of the area in each intensity and the $\eta_{\rm int}$ of 30% under macroscopic characterization indicates that the value of $\eta_{\rm int}$ fluctuates from approximately 10% to 50% within the active layer. It would be interesting to determine how this distribution changes with temperature, as well as with different LED samples. An investigation from this approach is now in progress.

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