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Spatial Inhomogeneity of Photoluminescence in InGaN Single Quantum Well Structures

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Spatial distribution of photoluminescence (PL) spectra has been assessed in an InGaN single quantum well (SQW) structure by means of fluorescence microscopy and scanning near-field optical microscopy (SNOM) under illumination-collection mode. The PL intensity of fluorescence image is uniform at 77 K, but the dark spot areas were extended with increasing temperature. The near-field PL images revealed the variation of both peak energy and intensity in PL spectra according to the probing location with the scale less than a few hundreds nm.

$In_xGa_{1-x}N$-based light emitting diodes (LEDs) are currently commercialized between the near-ultraviolet and amber spectral regions [1–3]. In spite of high threading dislocation density (10^8–10^{10} cm^{-2}), such LEDs exhibit a substantially high external quantum efficiency (η_{ext}) (10–15%) at the emission wavelength of blue region. These phenomena have been understood in terms of two major mechanisms. The first one is that the nonradiative recombination centers (NRC), whose main 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 fluctuation, so that the pathway to the NRC is hindered very effectively [4–6]. Nevertheless, it has also been reported that η_{ext} of InGaN-based LEDs decreases if the emission wavelength becomes longer than the blue-green region. Although the mechanism accountable for those phenomena has not been clarified yet, the key of this mechanism would be revealed by assessing the correlation between radiative/nonradiative recombination processes and micro/nanoscopic structures.

Several reports have recently appeared on spatial emission mapping measurements in InGaN single quantum wells (SQWs) by employing cathodoluminescence (CL) [7, 8] and scanning near-field optical microscopy (SNOM) [9–14]. 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 the spectroscopy. In this paper, the spatial inhomogeneity of photoluminescence has been investigated in an InGaN SQW structure by using the fluorescence microscope and SNOM under illumination-collection mode. The measurement of this mode leads to high spatial resolution because both photoexcitation and PL probing made by the same fiber tip prevent the spatial resolution from being affected by the diffusion effect of excitons and/or carriers [15].

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The sample used in this study was grown on (0001) oriented sapphire (Al_2O_3) substrate by the two-flow metalorganic chemical vapor deposition (MOCVD) technique [16]. The sample is composed of a GaN (1.5 μm), an n-GaN:Si (2.3 μm), an $In_{0.2}Ga_{0.8}N$ SQW (3 nm) and a GaN cap (5 nm) layer. The macroscopic PL peak of this sample was located at 470 nm at room temperature (RT). The fluorescent image was taken at temperatures from 77 K to RT using an optical microscope.

The near-field PL measurements were performed with an NFS-300 near-field spectrometer developed at JASCO Corp. The InGaN laser diode (LD, λ = 400 nm) was used as an excitation source in order to achieve the selective photo-excitation to an InGaN SQW. The optical power of 10 mW was coupled to the probe, and about 10 μW was utilized to illuminate the sample through the probe. PL collected by the probe (illumination-collection) was introduced into the 50 cm monochromator. The PL signal was detected by using a charge-coupled device (CCD) detector.

Figure 1 shows fluorescence images of a blue InGaN SQW taken in the range from 77 K to RT. The emission intensity was almost uniform at 77 K, the area as well as the density of dark spots were enhanced with increasing temperature. This is probably because the pathway to NRC was activated in addition to the increase of capture cross section to NRC [17].

Micro-PL and micro-time-resolved PL spectra of an InGaN SQW at 77 K and RT are shown in Figs. 2a and b, respectively. In each figure, PL spectra and decay profiles labeled (1)–(5) indicate the probing positions in the fluorescence image. The PL intensity and PL decay time fluctuate according to the location of the probing area reflecting that internal quantum efficiency of PL is inhomogeneously varied within the active layer. However, the detailed structures were blurred by the diffraction limit. Therefore, the PL mapping technique was developed using SNOM under an illumination-collection mode.

Near-field PL image of peak intensities and peak wavelengths were taken at an InGaN SQW structure by scanning 4 μm × 4 μm area with the mapping interval of 0.1 μm. The aperture size of the probe was about 0.3 μm in diameter. The spatial inhomogeneity with an area of a few hundreds nm was observed as shown in Fig. 3. Similar results have also been observed in other ternary alloys such as InGaAs [15] or GaNAs

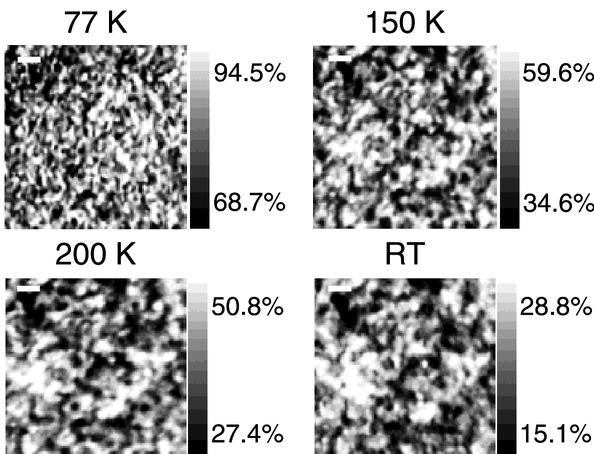

Fig. 1. The fluorescence images of an InGaN SQW at 77 K to RT. The white bars in the images indicate 5 μm scale

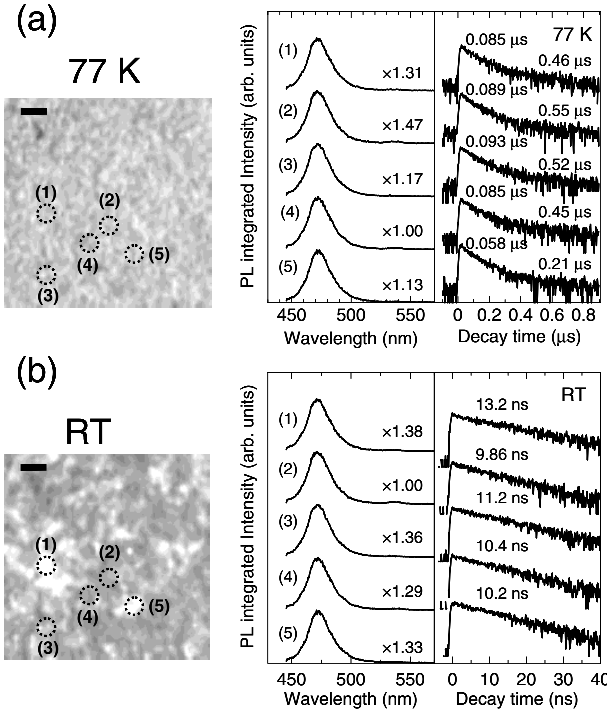

Fig. 2. Micro-PL and micro-time resolved PL spectra of InGaN SQW at a) 77 K and b) RT. The excitaion spot size and excitation power density are 2 μm × 2 μm and 9.375 μJ/cm^2, respectively. The bars in the fluorescence images indicate 5 μm. Dotted circles labelled with numbers indicate excitation positions in the fluorescence images

[18]. Spatial inhomogeneity of the PL integrated intensity indicate that the density of the defects and/or the NRC are distributed spatially. The results of Fig. 3 show a clear spatial correlation between PL integrated intensity and peak wavelength. The strong PL intensity regions correspond to long PL peak wavelength regions. These results suggest spatially inhomogeneous distribution of In alloy composition of the well layer. It would be interesting how such spatially inhomogeneous distribution changes with temperature as well as In composition of well layer of various SQW-LED samples. Such approach is now in progress.

We have studied a spatial inhomogeneity of PL in an InGaN SQW structure using fluorescence microscopy and SNOM under illumination-collection mode. The emission intensity of fluorescence images was almost uniform at 77 K, the area as well as the

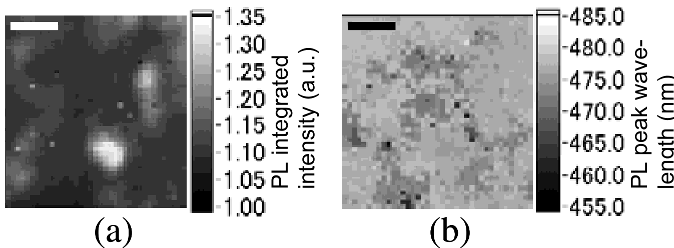

Fig. 3. a) Near-field PL image mapped with the PL integrated intensity and b) peak wavelength at RT. The bars in the images indicate 1 μm scale. The excitation power density is 1.11 × 10^3 W/cm^2 under continuous wave (cw) condition

density of dark spots were enhanced with increasing temperature. The near-field PL mapping image obtained in this investigation revealed the variation of both peak and intensity in PL spectra according to the probing location with the area less than a few hundreds nm.

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