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Phase Separation Mechanism Around Dislocation in an InGaN/GaN Quantum Well Structure

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Abstract. The role of dislocation for luminescence in InGaN grown on sapphire substrate by metal organic chemical vapor deposition (MOCVD) method was investigated by cathodoluminescence (CL), transmission electron microscopy (TEM) and atomic force microscopy (AFM). The CL emission area and dark spots between InGaN and GaN layers in InGaN/GaN single quantum well (SQW) and multiple quantum well (MQW) structures showed completely one to one correspondence indicating that dislocations in InGaN act as non-radiative recombination centers. It was confirmed that the phase separation in InGaN occurs when InGaN layer is thick and the growth temperature is low. The phase separation growth mechanism which is caused by spiral growth due to mixed dislocations was discussed.

1. Introduction

GaN based blue-to-green light emitting diodes (LEDs) with InGaN active layer have been already developed and commercialized[1]-[2], and a laser diode (LD) has also been developed.[3]-[4] However, in spite of a lot of dislocations (density of 10^8-10^{11} cm^{-2}) in InGaN layer produced by hetero-epitaxy with a sapphire substrate[5], the light emission efficiency of these devices is very high. From this reason it is important for the high performance device to clarify the role of dislocation. Recently we reported that dislocations work as non-radiative recombination centers in GaN[6] and proposed the InGaN phase separation model formed by different indium incorporation rate at the dislocation site.[7] Keller *et al.* reported that the dislocations strongly influence the growth mechanism and the morphology of InGaN SQWs and bulk films[8]. More recently, on the other hand, Mukai *et al.* reported using LEDs on sapphire and on epitaxially laterally overgrown GaN (ELOG) that the dislocations do not act as non-radiative recombination centers in InGaN, and the indium fluctuation is not caused by dislocations in InGaN.[9] This paper is misleading in two terms. First, they did not distinguish "phase separation" and "compositional fluctuation" of InGaN layers. The former is produced by the dislocation as described in our previous and in the present papers, however, the latter is available in most of the ternary and quaternary alloys[10]-[11] including InGaN grown on dislocation-free bulk GaN. Second, they ruled out the possibility of very short diffusion length to explain high quantum efficiency of their LEDs fabricated on a dislocated GaN on sapphire.

In this paper, using spatially resolved cathodoluminescence (CL) images we show that dislocations act as non-radiative recombination centers in InGaN, and that the phase separation into high- and low- indium compositional InGaN takes place when InGaN growth temperature is low and its thickness exceeds some critical value. The phase separation mechanisms and a possible reason for discrepancy between our results and ref. [9] is also discussed.

2. Experiment

The InGaN/GaN single quantum well (SQW) and multiple quantum well (MQW) were grown on sapphire (0001) substrate by horizontal atmospheric metal organic chemical vapor deposition (MOCVD) system.[12] Flow rate of trimethylgallium (TMG), trimethylindium (TMI) and NH_3 used as source gases in InGaN growth were 44 μmol/min, 116 μmol/min, and 10 SLM, respectively. The structure of four samples prepared in this study is GaN (0.1 μm)/(i), (ii), (iii) or (iv)/GaN(1.5 μm)/buffer GaN (2.5 nm)/sapphire(0001). [(i) $In_{0.126}Ga_{0.874}N$(2.5 nm) SQW grown at 715℃, (ii) $In_{0.126}Ga_{0.874}N$(2.5 nm)/GaN(7.5 nm) 10 MQW grown at 715℃, (iii) $In_{0.140}Ga_{0.860}N$(2.2 nm)/GaN(6.0 nm) 10 MQW grown at 675℃, and (iv) $In_{0.126}Ga_{0.874}N$(5.0 nm)/GaN(7.5nm) MQW grown at 715℃] Note that {(i), (ii)}, {(ii), (iv)}, and {(ii), (iii)}, respectively, are different only in the number, the thickness and the growth temperature of the quantum wells. By decreasing the growth temperature from 715 to 675 ℃, indium composition is increased and the quantum well thickness was decreased due to the decreased growth rate. The growth temperatures for GaN buffer layer and GaN epitaxial layer are 450 and 1050℃, respectively. All layers are undoped.

The formation of MQW periodic structure was confirmed by cross-sectional transmission electron microscopy (TEM) pictures and satellite peaks of X-ray diffraction spectra. The film thickness and the indium composition were determined by TEM measurement and X-ray spectra, respectively. CL measurements were performed with an acceleration voltage of 5 or 15 kV using a JEOL 6400 scanning electron microscope equipped with an Oxford Instrument Mono CL2. For GaN, the maximum penetration depth of an electron beam has a calculated value of < 0.3 μm for 5 keV, <1.3 μm for 15keV.[13][14] Plan-view and cross-sectional CL images were taken using as-grown and thinned TEM sample, respectively. Atomic force microscopy (AFM) image were taken by non-contact mode using a Seiko Instrument SPA300.

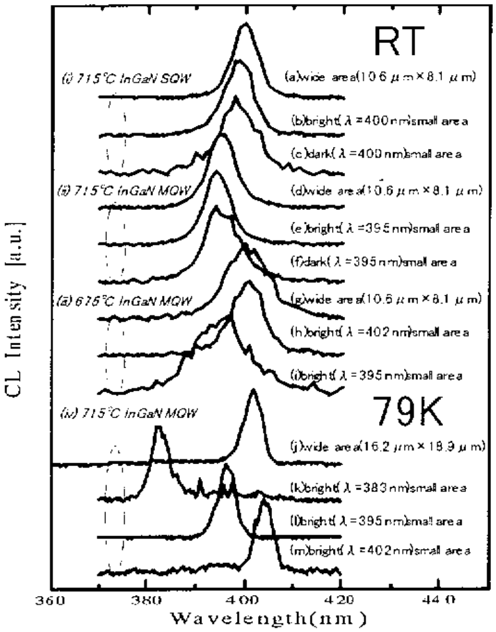

Fig.1 RT or 79K CL spectra for sample (i), (ii), (iii) and (iv). Small area measurements are about 100-200nm in diameter at the different points

3. Results and Discussions

Figure 1 shows respectively room temperature or 79K CL spectra which are taken by scanning incident electron beam in relatively wide area and by fixing electron beam at certain points on the samples. The small area of 100-200nm in diameter spectra are taken at two or three different position in the same sample. Although the peak from GaN is also visible at around 361 nm in all of the four samples, its peak intensity is approximately three orders of magnitude smaller than those from the SQW or MQW.

The peak position and the half width of three spectra of the sample (i) (SQW grown at 715℃) are almost the same. The same is true also for sample (ii) (MQW grown at 715℃). However, the wide area spectra of sample (iii) (2.2 nm MQW grown at 675℃) and (iv) (5 nm MQW grown at 715℃) are wider than those of the small area spectrum, and the peak wavelengths of the small area spectra are different at two or three different points. These indicate that InGaN layers in samples (iii) and (iv) are phase separated into high indium and low indium regions.

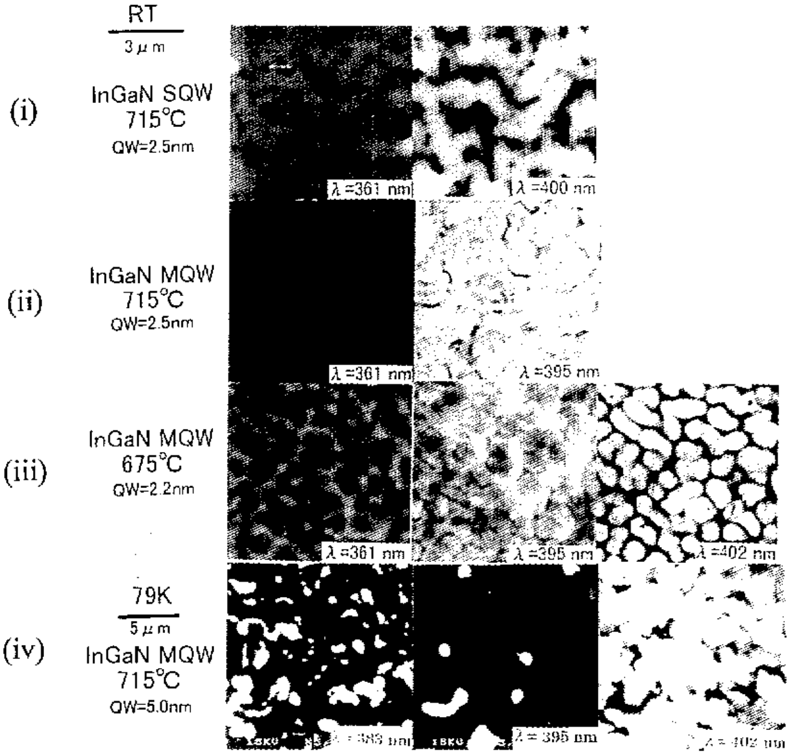

Fig. 2 Room temperature or 79K plan-view CL images for sample (i), (ii), (iii) and (iv) taken at the same location.

Figure 2 shows plan-view CL images taken at wavelength corresponding to the InGaN and the GaN band edge (λ =361 nm) emissions of each sample at RT, and shows plan-view 79K-CL images of sample (iv) taken at different three wavelengths of the InGaN emissions. The surfaces of all samples are smooth and featureless, but a high density of dark spots of the order of 10^8-10^9 cm^{-2} are observed in all CL images except those of sample (iv), and there are one to one correspondence between the dark spots taken at InGaN and GaN wavelengths (see, for example, InGaN SQW grown at 715℃ in Fig. 2). From our previous results[6] the one to one correspondence between CL dark spots and threading dislocations in GaN is very clear. Hence the correlation between the InGaN and GaN dark spots clearly demonstrates that threading dislocations are appearing as dark spots in InGaN. This conclusion, a dislocation being a non-radiative recombination center in InGaN, contradicts to ref. [9] which shows no difference in emission efficiencies of LEDs fabricated on dislocated GaN on sapphire and low dislocation ELOG substrates. It should be pointed out that the CL dark spots were also observed in their materials.[15] Therefore, two explanations may be possible to explain this discrepancy. One possibility is a very short minority carrier diffusion length in InGaN. If minority carriers recombine radiatively before they are captured by the dislocation, the emission efficiency can be high. Due to our formula[6] and assuming minority carrier diffusion length of 10 nm, the emission efficiency is 0.95 even at dislocation density of 10^{10} cm^{-2}. The second possibility is the difference between the current injection and the CL. The current path through InGaN layer may not be uniform, and if there is a mechanism that the current avoids dislocation to flow, the LED emission does not degrade even with high density non-radiative centers.

It is visible for samples (i) and (ii) (SQW and MQW grown at 715℃) that both CL images of InGaN show very homogeneous emission pattern except for a lot of dark spots, therefore InGaN MQW emission pattern can be understood as an overlapped image of InGaN SQW. This also means that dark spots locate at the same points in each of the 10 InGaN layers in MQW, because the dark spot densities are almost equal for MQW and for SQW. On the other hand, the CL images of sample (iii) taken at two different wavelengths are completely complementary. This means that the InGaN layer is separated into two phases; one is high indium content (λ =402 nm) and the other is low indium (λ =395 nm). In addition to this phase separation, many dark spots randomly distribute on the surface are visible in bright area of both 402 nm and 395 nm CL images. These small dark pits correspond to the dislocations.

It was also found by high magnification CL as shown in Fig.3 (a) that a pair of small dark spots exists in all of the bright area of 402 nm CL image. These pairs of pits were also found in AFM image as shown in Fig.3 (b) for the same sample and were explained as threading dislocations with screw components[8]. Therefore, the bright areas of 402 nm CL (higher indium domain) are formed by the dislocation with screw component.

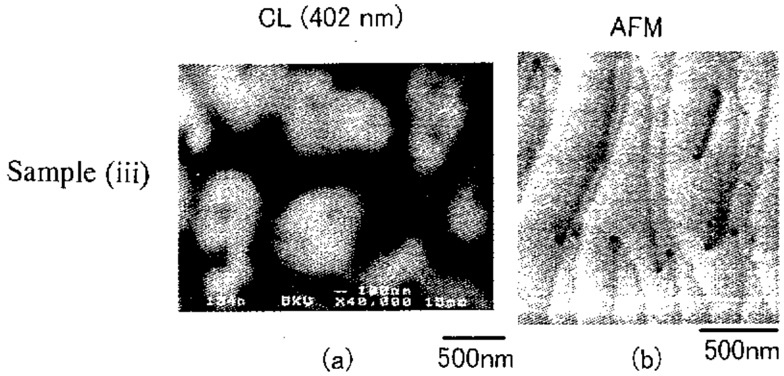

Fig.3 (a) : A room temperature-plan-view high magnification 402 nm CL images. (b) : an AFM image of top GaN surface. Both image were taken for sample (iii) ($In_{0.140}Ga_{0.860}N$ MQW grown at 675℃).

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Figure 4 is the cross sectional CL images of sample (ii) at 79 K taken at three different wavelengths of 388, 397 and 357 nm (GaN band edge). These pictures are taken at exactly the same position. The dark region marked by dashed line in GaN CL corresponds threading dislocation in GaN. CL at 397 nm (high indium) and 388 nm (low indium) are bright and dark, respectively at this dislocation site. This is another evidence that indium composition is higher at dislocation site.

The phase separated InGaN layer may be formed by the spiral growth nucleated at the dislocation with screw component. These nuclei coalesce with growth, and the bigger nuclei overcome the smaller, otherwise the domain size of the higher indium region does not become so uniform as shown in Fig.2 (CL at 402nm) considering dislocations distribute randomly on the GaN surface.

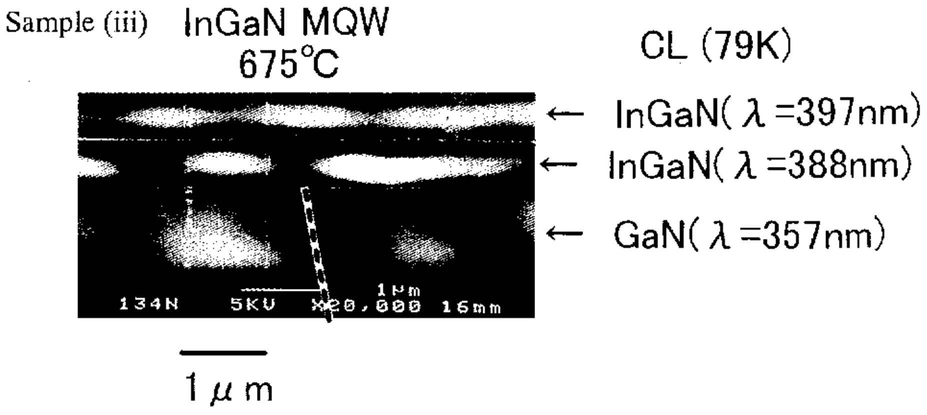

Fig.4 79K-cross-sectional CL images of sample (iii) ($In_{0.140}Ga_{0.860}N$ MQW grown at 675℃). Both InGaN and GaN CL images are at the same location. The dark region marked by the dashed line in GaN CL corresponds threading dislocation in GaN.

According to the spiral model developed by Burton, Cabrera and Frank (BCF Model)[16], the driving force $\Delta\mu$ to grow a crystal is given by

$$\Delta\mu \sim \frac{\Omega\beta^2}{k_B T}$$

where Ω and k_B are the area per one molecule and Boltzmann constant, respectively. And β (the free energy per unit length of a step) increases with decreasing temperature. The growth velocity of adhesive mode is linear to $\Delta\mu$, but that of the spiral mode is linear to $(\Delta\mu)^2$. Therefore if the growth temperature in InGaN decreases, spiral growth should be more dominant. The phase separation takes place only at low growth temperatures, and the dislocation triggers nucleation of spiral growth.

This conclusion is consistent with our previous results[7] in which a big difference in indium inhomogeneity is found between InGaN layers grown on a dislocation-free homoepitaxial GaN and on a dislocated GaN on sapphire. The assumption of higher indium incorporation rate at the dislocation site is reasonable, if the indium evaporation which is always happening at the InGaN growth temperature is taken into account. Even if the deposition rate is uniform over the entire surface, the evaporation rate at the dislocation site must be slower due to more dangling bonds available at the dislocation site.

In Fig.2 it can be visible that InGaN phases complementarily separate with three regions (λ =383, 395, 402 nm) in spite that this MQW is grown at 715℃ and also that the emission of about 70-80 % is λ =402 nm. The MQWs grown under the same conditions with InGaN thickness of 1.25 nm (data not shown here) and 2.5 nm (sample (ii)) showed no phase separation even at growth temperature of 715℃. Therefore, only growth tempewrature cannot explain this phenomenon. Followings are possible phase separation mechanisms to explain these experimental results. 1. Strain relaxation and dislocation generation in InGaN when its thickness exceeds critical layer thickness. 2. Indium diffusion effect due to longer growth time. One or more in above may be responsible.

4. Summary

Role of dislocation in InGaN SQW and MQW structures were investigated by cathodoluminescence (CL) and atomic force microscopy (AFM). Two conclusions can be made. First, the dislocation in InGaN is a non-radiative recombination center as long as we characterize materials by CL. A possible reason of the discrepancy of this conclusion and that of ref [9] is discussed. Second, a phase separation in a InGaN layer into higher and lower indium regions occurs when growth temperature is low and InGaN is thicker than critical value. In any case, the dislocation with screw component is helping phase separation to happen. The SQW and MQW with thin InGaN grown at 715℃ is uniform even with the existence of the dislocation. However the compositional fluctuation may exist, because the CL spectrum half width from the InGaN layer is much broader than that from GaN.

Acknowledgments

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