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The enhancement of phase separation in the InGaN layer grown on a GaN layer with a rough surface was investigated for the formation of self-assembled In-rich quantum dots (QDs) in the InGaN layer. Transmission electron microscopy images showed that In-rich QDs with a size of 2–5 nm were formed even in an InGaN layer with a low indium content, and a layer thickness less than the critical thickness. The room-temperature photoluminescence (PL) spectrum of this layer showed emission peaks corresponding to In-rich QDs. The temperature-dependent PL spectra showed dominant peak shifts to the lower energy side, indicating that the self-assembled In-rich QDs are formed in the InGaN layer grown on a rough GaN surface and that the carriers are localized in In-rich QDs. © *2005 American Institute of Physics.* [DOI: 10.1063/1.2008365]

InGaN alloys have recently attracted attention as potential materials for active layers in high efficiency light-emitting diodes (LEDs) and laser diodes operating in the visible and ultraviolet wavelength range.[1] It has been proposed that the high luminescence efficiency results from self-formed In-rich regions in spite of the large defect density in the InGaN layers.[2–6] These In-rich regions are believed to act as In-rich quantum dots (QDs) in the InGaN layer, providing deep potential wells that suppress the diffusion of electrical carriers toward various nonradiative defects. The carrier localization within In-rich QDs in the InGaN layer also has a significant effect on the performance of LEDs, resulting in a large redshift in the emission energy and an increase in radiative recombination efficiencies.[7] In-rich QDs in InGaN are formed by a compositional fluctuation or phase separation. The compositional fluctuation of indium is commonly observed in the majority of InGaN alloys, but phase separation can occur in InGaN layers with high In contents, due to the low miscibility of InN in GaN. Theoretical calculations predict that phase separation in an InGaN layer occurs below the critical temperature and for a range of composition of the alloy that defines a miscibility gap at a given growth temperature.[8] However, phase separation is kinetically inhibited, due to the nonequilibrium state associated with the conventional growth condition. Furthermore, phase separation in an InGaN alloy on the GaN layer has been reported to be suppressed by biaxial compressive strain due to the large lattice mismatch of InGaN to the underlying GaN layer.[9,10] As a result, In-rich QDs formed by phase separation have been observed in InGaN films that contain high concentrations of In,[11,12] a layer thickness larger than the critical layer thickness,[9,13] or a layer that was thermally annealed at high temperature.[14,15] As the thickness or the In composition in InGaN layer increases, however, the driving force for phase separation increases, but the crystalline quality of InGaN alloys deteriorates drastically. Therefore, it would be desirable to induce phase separation to form the In-rich QDs even in an InGaN layer with a low In concentration or a thickness below the critical layer thickness without the postgrowth thermal process.

In this paper, we report on the growth of self-assembled In-rich QDs by enhanced phase separation in the InGaN layer, which was grown on a rough GaN layer. Strain in the InGaN layer could be relieved by introducing a rough GaN surface as an underlying layer, and consequently, the In-rich QDs were spontaneously formed in the InGaN layer.

The $In_{0.16}Ga_{0.84}N$ films were grown on (0001) *c*-plane sapphire substrates at a pressure of 200 Torr by metal-organic chemical-vapor deposition (MOCVD). After annealing the substrate in H_2 at 1050 °C, a 25-nm-thick GaN buffer layer was deposited at 550 °C, followed by the deposition of a 2-µm-thick undoped GaN layer grown at 1050 °C. The temperature was then decreased to 750 °C, and a 30-nm-thick $In_{0.16}Ga_{0.84}N$ layer was grown on the GaN layer. In order to investigate the effect of the surface roughness of the GaN layer on phase separation in the InGaN layer, InGaN films were grown on both smooth and rough GaN surfaces. Rough GaN surfaces were prepared by supplying trimethyl gallium (TMGa) source gas during the period when the growth temperature was lowered, prior to InGaN growth.

The surface morphology of the GaN surface was measured by atomic force microscopy (AFM) in the contact mode. Figures 1(a) and 1(b) show AFM images of the flat

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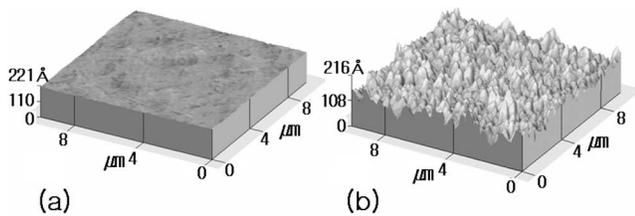
FIG. 1. AFM images of surface morphologies of (a) a smooth GaN with a rms roughness of 2.5 Å and (b) a rough GaN with a rms roughness of 23.5 Å.

and rough GaN surfaces with a root mean square (rms) roughness of 2.5 and 23.5 Å, respectively. The AFM images show that the surface roughness of the GaN is increased as the result of supplying TMGa source gas during the period when the temperature is ramped down from the growth temperature of GaN at 1050 °C to that of InGaN at 750 °C. Surface roughening would be expected on the GaN layer as shown in Fig. 1(b), since the surface diffusion coefficient of Ga adatoms is exponentially decreased with decreasing surface temperature.[16]

Figures 2(a) and 2(b) show the cross-sectional transmission electron microscopy (TEM) images of InGaN layers grown on smooth and rough GaN surfaces, respectively. As shown in Fig. 2(a), the InGaN layer grown on a flat GaN surface shows that the interface between InGaN and GaN layers is abrupt and there is no evidence of a phase separation in the InGaN layer. The critical layer thickness of the $In_{0.16}Ga_{0.84}N$/GaN bilayer structure was estimated to be 50 nm, based on the energy balance model.[17,18] Because the thickness of the InGaN layer in this study was 30 nm, the InGaN layer is assumed to be pseudomorphically grown on the GaN under compressive strain. Therefore, phase separation would not be expected to occur due to the coherence strain,[9] even though the In composition of 16% is in the miscibility gap for InGaN. This agrees with the results of Fig. 2(a), which shows that the creation of dislocations or the formation of the phase-separated In-rich QDs did not occur in the InGaN layer, since the compressive strain energy is not released.[13] However, in the case of the InGaN layer grown on a rough GaN surface, numerous dark spots are observed across the InGaN layer as shown in the inset of Fig. 2(b). The dark regions in the TEM image of the InGaN layer can be attributed to phase-separated In-rich QDs.[2,3,6,7,11,13] The diameter of these In-rich QDs is in a range from 2 to 5 nm. These results indicate that the rough GaN layer induces phase separation in InGaN film even though the thickness of the InGaN layer is less than the critical layer thickness and the InGaN with a low In composition. In incorporation in InGaN film can be increased to form In-rich QDs by decreas-
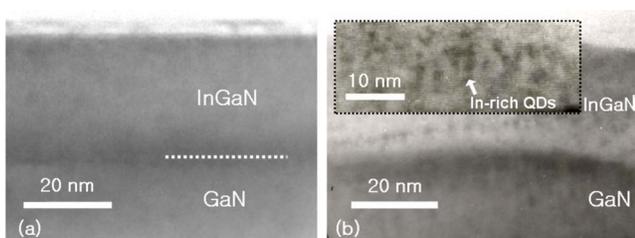
FIG. 2. Cross-sectional TEM images of InGaN layers (a) grown on a smooth GaN surface and (b) grown on a rough GaN surface. The inset of (b) shows In-rich QD regions in InGaN grown on a rough GaN surface.
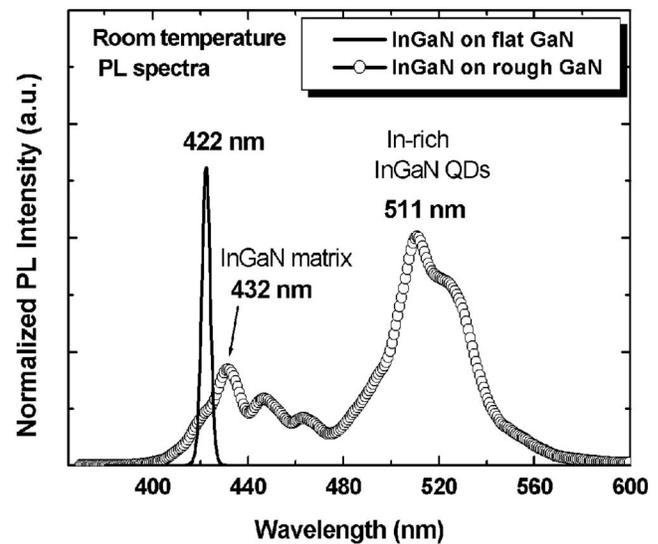
FIG. 3. Room-temperature PL spectra of InGaN layers grown on the smooth and rough GaN layers.

ing the strain-induced composition pulling effects that would be caused by compressive strain in the InGaN layer.[19] However, x-ray diffraction results showed the same In composition of 16% for the InGaN layers grown on both smooth and rough GaN surfaces. From these results, the rough GaN surface layer is believed to play a critical role in phase separation by relieving the compressive strain in the InGaN film, thus permitting self-assembled In-rich QDs to be formed in the InGaN layer.

A photoluminescence (PL) measurement was carried out on the InGaN films to investigate the optical properties. Figure 3 shows the room-temperature PL spectra of InGaN layers grown on both smooth and rough GaN surfaces. A strong emission peak at 422 nm (2.94 eV) was observed on the InGaN layer grown on a smooth GaN surface. The PL peak position at 422 nm is blueshifted by about 0.14 eV compared to the PL peak position of 2.80 eV (442 nm), which corresponds to a thick InGaN film with an In mole fraction of 0.16.[20] This can be explained by the increase in the band gap of the InGaN layer, which was pseudomorphically grown on the flat GaN layer under biaxial compressive strain.[21] This result is consistent with the TEM images shown in Fig. 2(a), which shows that strain relaxation does not occur in the InGaN layer grown on a smooth GaN surface. On the other hand, the PL spectrum of the InGaN layer grown on a rough GaN surface shows broad PL peaks at around 432 and 511 nm, which can be attributed to the InGaN matrix and the In-rich QDs, respectively. The higher energy PL peak at 432 nm (2.87 eV) is redshifted compared to the PL peak of the InGaN layer grown on a smooth GaN layer. This result is attributed to the strain relaxation in the InGaN layer due to the underlying rough GaN, which can cause a redshift in PL peak of InGaN matrix. The PL peak at 511 nm is not believed to originate from the thickness fluctuation of the InGaN layer but from the In-rich QDs because the thickness of InGaN layer is too thick to show a quantum size effect. The redshift in the PL peaks can be attributed to the relief of compressive strain in the InGaN layer and the formation of phase-separated In-rich QDs on the rough GaN surface.

The temperature-dependent PL spectra were obtained for an InGaN layer grown on a rough GaN surface as shown in
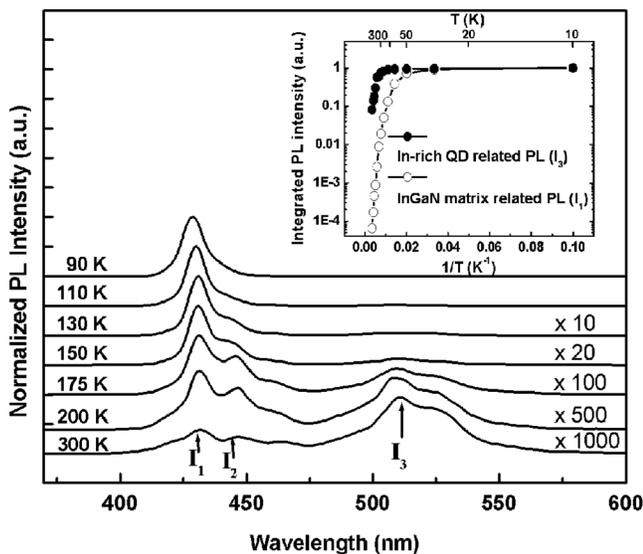
FIG. 4. Temperature-dependent PL spectra of an InGaN layer grown on a rough GaN surface. The inset shows normalized integrated PL intensity as a function of $1/T$ for In-rich QDs related emission (closed circle) and InGaN matrix related emission (open circle).

Fig. 4. At a temperature of 90 K, the band-edge related peak of the InGaN matrix is observed at 430 nm. As the temperature increases, the intensity of peak I_1 rapidly decreases and a low energy tail peak, labeled I_2, begins to appear and is strongly developed at a high temperature of 175 K. With a further increase in temperature above 200 K, electron-phonon scattering becomes important, resulting in the broadening and decay of peaks I_1 and I_2. It should be noted that another In-rich QD related peak, labeled I_3, dominates at temperatures above 200 K. As shown in Fig. 4, the dominant peak is changed from I_1 to I_3 as the temperature increases. This result can be attributed to the thermally activated carrier transfer process between neighboring In-rich QDs, which were formed by phase separation in the InGaN layer.[13,22]

The inset of Fig. 4 shows Arrhenius plots of the normalized integrated PL intensity for the InGaN matrix related PL peak (I_1) and In-rich QD related peak (I_3). The PL intensity rapidly decreases above 50 K for I_1 and above 90 K for I_3. This indicates that excitons in the In-rich QDs are in deeper confining potentials than those in the InGaN matrix, since the quenching of the PL intensity with temperature can be explained by the thermal emission of carriers out of confining potentials. In addition, as the temperature increases from 10 to 300 K, the integrated PL intensities are decreased by an order of magnitude of 1 for I_3 and 4 for I_1 peak. This result also indicates that the carriers transfer from the thermally unstable higher potential state in the InGaN matrix to the thermally stable lower potential states in the In-rich QDs with increasing temperature. The TEM and PL results show that the phase separation in the InGaN layer with a low In concentration can be promoted by growing the InGaN layer on a rough GaN layer to spontaneously form the In-rich QDs in the InGaN layer.

In summary, the formation of phase-separated In-rich QDs formed in the InGaN layer with a low In concentration and a thickness smaller than the critical thickness was investigated. Cross-sectional TEM images showed that In-rich QDs with a size of 2–5 nm were formed in an InGaN layer grown on a rough GaN surface by relieving the strain in the InGaN layer. The temperature-dependent PL spectra also showed that self-assembled In-rich QDs with thermally stable and deeply localized states can be formed, even in InGaN layers with a low In concentration by the surface roughness of the underlying GaN surface.

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