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Green Gap Spectral Range Light-Emitting Diodes with Self-Assembled InGaN Quantum Dots Formed by Enhanced Phase Separation

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Light-emitting diodes (LEDs) emitting in the green gap spectral range (540–610 nm) were demonstrated using self-assembled In-rich InGaN quantum dots (QDs) grown on n-GaN by metal-organic chemical-vapor deposition. The study of structural and optical properties showed that the formation of InGaN QDs with larger size and higher In composition is enhanced by phase separation in the InGaN layer with increasing surface roughness of the underlying n-GaN layer. The emission wavelength of the LED was redshifted from the green (540 nm) to red (610 nm) spectral range due to an increase in the depth of potential wells of the InGaN QDs with increasing surface roughness of the underlying n-GaN layer.
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Group III–nitride-based light-emitting diodes (LEDs) have been rapidly developed for use in solid-state lighting applications.[1,2] In particular, white LEDs have attracted considerable attention not only for the general lighting but also for back light units in display applications. Although conventional white LEDs use the phosphor conversion method to produce white light from a blue or ultraviolet LED,[1] having good green and red sources could lead to color-mixed white sources that would avoid the losses associated with this approach. Blue LEDs based on InGaN/GaN multiple quantum wells (MQWs) are now commercially available, but the spectral range between 550 and 590 nm is the well known "green gap" in which the efficiency of InGaN-based LEDs decreases significantly.[3–5] The InGaN/GaN MQWs with In composition more than 20% can suffer from the formation of defects that leads to low radiative efficiency.[4] Furthermore, the incorporation of In atoms into InGaN is limited due to a compositional pulling effect caused by a large lattice mismatch between InGaN and GaN.[6,7] To circumvent the problems inherent to the InGaN MQW approaches, alternate active layers such as InGaN nanorods[8,9] and quantum dot (QD)[1,4,10] structures have been suggested as light emitters in the green gap spectral range. However, there is no report on controlling the emission wavelength of the LEDs in this spectral range. In this paper, we report on the enhanced phase separation in the InGaN layers to form the In-rich InGaN QDs for the fabrication of LEDs that emit in the green gap wavelength spectral range.

Self-assembled In-rich InGaN QDs were found to be spontaneously formed in a InGaN layer grown on a rough GaN layer on a (0001) *c*-plane sapphire substrate at a pressure of 200 Torr in a metalorganic chemical vapor deposition (MOCVD) system.[11] After annealing the sapphire substrate in H_2 at 1050 °C, a 25-nm-thick GaN buffer layer was deposited at 550 °C, followed by deposition of a 1-μm-thick undoped GaN layer and a 2-μm-thick n-GaN:Si layer at 1050 °C. A rough n-GaN:Si layer was formed by supplying the source gases while reducing the growth temperature prior to the growth of an InGaN active layer. The surface roughness was controlled by changing the growth time of the rough n-GaN:Si. An InGaN layer with a thickness of 30 nm was grown at 750 °C on n-GaN:Si layers with

different surface roughnesses. To grow LED structures, a 200-nm-thick Mg-doped p-GaN layer was grown on the active InGaN layer at 900 °C. The detailed procedure for the LEDs with a size of $300 \times 300 \, \mu m^2$ was reported elsewhere.[4]

Figures 1(a)–1(d) show the evolution of surface morphologies of the n-GaN surfaces with increasing growth time from 0 to 6 min. The surface morphologies were observed by atomic force microscopy (AFM). A growth time of 0 min means that the source gases are not supplied while reducing the growth temperature from the growth temperature of n-GaN to that of an InGaN active layer. When the growth time of the rough n-GaN surface was 0 min, the surface atomic structure was well defined, but the surfaces became rough with increasing growth time of the rough n-GaN surface. As shown in Fig. 1(e), the root mean square (rms) roughness of the n-GaN surface increased from 2.67 to 36.2 Å as the growth time increased from 0 to 6 min. This result can be explained by a model for surface diffusion based on an Einstein relation:[12]

$$x^2 = 2D\tau, \tag{1}$$

where x is the mean displacement distance, D the diffusion coefficient, and τ the average diffusion time. The diffusion coefficient D is a function of temperature, as shown by[12]

$$D = D_0 \exp\left(-\frac{E_D}{kT}\right), \tag{2}$$

where D_0 is a constant, E_D is the activation energy of surface diffusion for surface atoms, k is Boltzmann's constant, and T is the temperature of the growth surface. These equations show that the surface diffusion coefficient decreases exponentially with decreasing surface temperature, finally resulting in a reduction of the diffusion distance of source adatoms on the growth surface. Therefore, as the growth time of the n-GaN surface increases while reducing the growth temperature, the surface roughness increases due to the decreased diffusion distance of Ga adatoms.

Figure 2(a) shows normalized θ–2θ X-ray diffraction (XRD) patterns for InGaN layers grown on n-GaN layers with different surface roughnesses. As the surface roughness of the underlying n-GaN layer increases, the peaks from the InGaN layer broaden, while the peak position remains fixed. The In composition and the full width at half maximum (FWHM) of a peak from the InGaN layer are plotted as shown in Fig. 2(b). As shown in Fig. 2(b), the FWHM
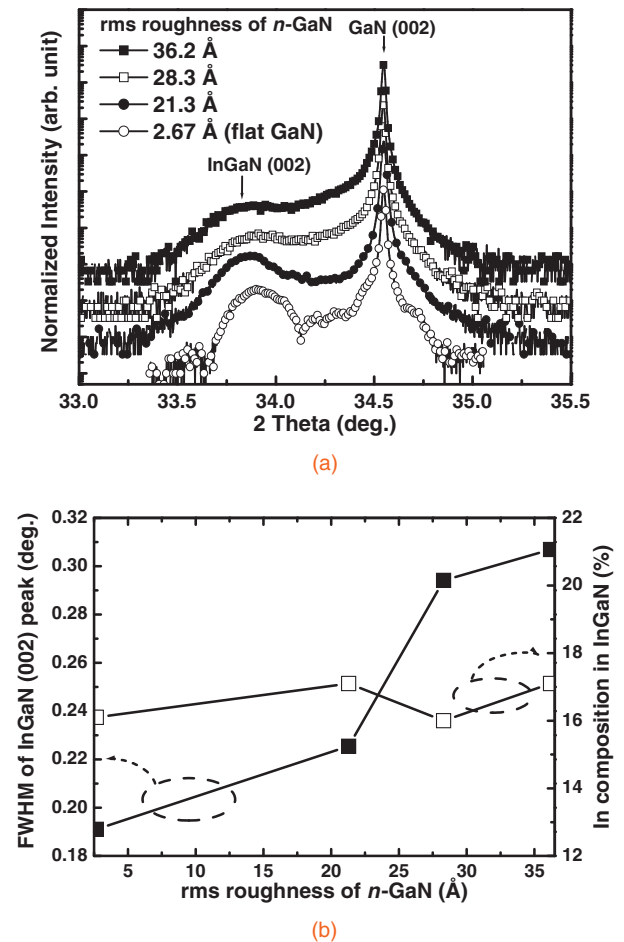
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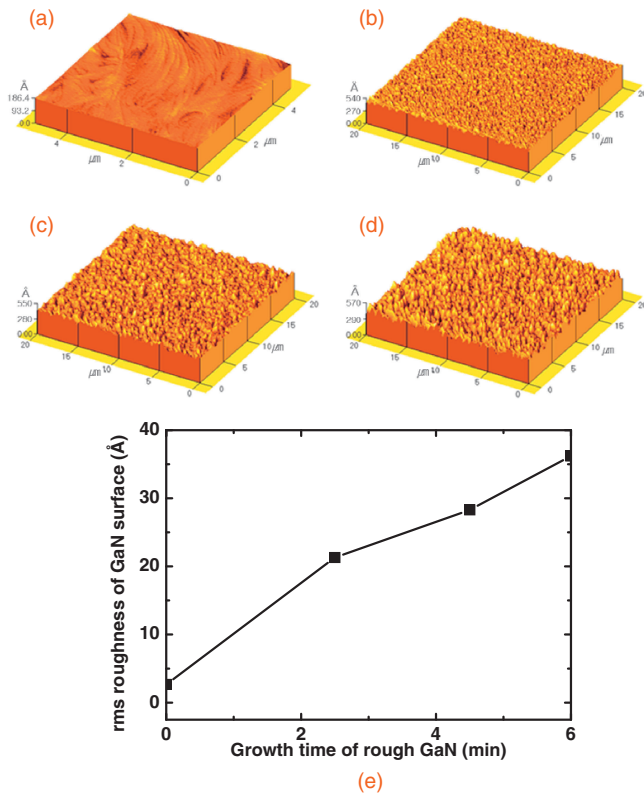
Fig. 1. AFM images of surface morphologies of GaN layers with different rms roughnesses of (a) 2.67, (b) 21.3, (c) 28.3, and (d) 36.2 Å. (e) RMS roughness of GaN surface versus the growth time of rough n-GaN.

Fig. 2. (a) θ–2θ XRD scan spectra of InGaN layer in the (002) reflection grown on rough GaN. (b) FWHM of the InGaN (002) reflection and In composition in InGaN vs the rms roughness of n-GaN.

Fig. 3. Normalized PL spectra of InGaN layers grown on rough GaN layers with different rms roughnesses measured at room temperature. The multiple Gaussian peaks for the deconvolution of the PL spectrum are shown by dashed lines.

increases from 0.19 to 0.31 degrees with increasing surface roughness of the n-GaN layer from 2.67 to 36.2 Å, while the In composition slightly varies between 16 and 17%. The broadening of InGaN peaks is the combined result of alloy fluctuation and finite domain size effects.[13,14] This indicates that alloy fluctuation and formation of nanometer size domains in the InGaN layer are enhanced by increasing the surface roughness of the underlying n-GaN layer. It is known that the formation of In-rich InGaN QDs due to the phase separation is enhanced when compressive stress in the InGaN film is relaxed by the underlying GaN layer.[11,15] Therefore, it is believed that the formation of In-rich InGaN QDs in the InGaN layer is accelerated by increasing the surface roughness of the underlying n-GaN layer because of further relaxation of the compressive stress in the InGaN layers.

Figure 3 shows photoluminescence (PL) spectra of the InGaN layers grown on the n-GaN layers with different surface roughnesses. As shown in Fig. 3, an InGaN layer grown on n-GaN with a flat surface shows a single emission peak at 420 nm (2.95 eV), which is blueshifted from the PL peak at 442 nm (2.80 eV) of a thick InGaN film with an In mole fraction of 0.16 due to a biaxial compressive strain. By increasing the surface roughness of the underlying n-GaN layer from 21.3 to 36.2 Å, the spectral lines of the PL spectra change from fivefold to double peaks, as shown in Fig. 3. The PL spectra were fitted by a multiple Gaussian. The broad PL spectrum for the InGaN layer on n-GaN with an rms roughness of 21.3 Å is composed of PL peaks at 459, 497, 529, 565, and 591 nm as noted by dashed lines (P1–P5). The PL spectrum shows PL peaks in the short wavelength (P1 and P2) and long wavelength (P3–P5) regions. The PL
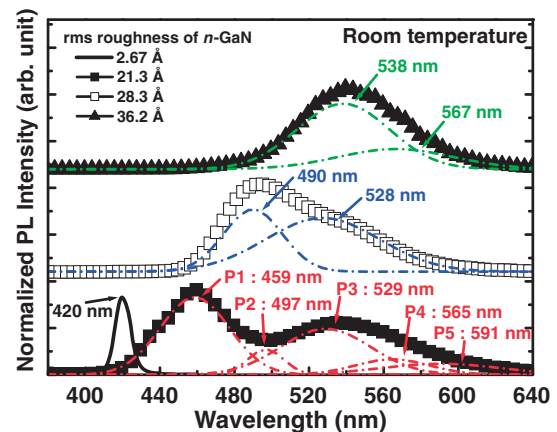
peaks in the short and long wavelength regions are attributed to the emissions from the In-poor InGaN matrix and In-rich InGaN QD layers formed by enhanced phase separation, respectively. The broad PL emission from the In-poor InGaN matrix layer can be due to the inhomogeneity of In composition and stress in the InGaN layer. Three peaks in the long wavelength region, P3–P5, are obtained from In-
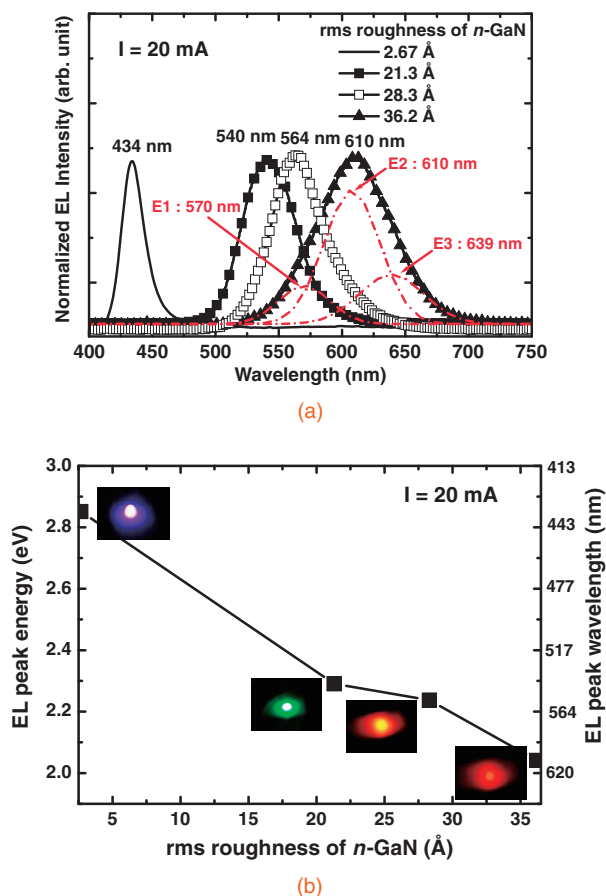
Fig. 4. (a) Normalized EL spectra of In-rich InGaN QD LEDs grown on rough GaN surfaces with different rms roughnesses. Three elementary EL spectra shown by dashed lines (E1–E3) are obtained by deconvoluting the EL spectrum of an LED with a roughness of 36.2 Å. (b) Peak position and the FWHM of EL peak versus the rms roughness of n-GaN. The inset shows the EL images of InGaN QD LEDs measured at an input current of 20 mA.

rich InGaN QD ensembles with different distributions of In composition and size. As the surface roughness of the n-GaN layer is further increased, the PL spectra are dominated by the long wavelength peaks corresponding to the In-rich InGaN QD ensemble with large size and high In content, which provide deep potential wells. The PL results show that the phase-separated InGaN layers in this study contain three different InGaN QD ensembles corresponding to P3–P5, but the fraction of QD ensembles changes with the surface roughness of the underlying n-GaN layer.

Figures 4(a) and 4(b) show the electroluminescence (EL) results of LEDs with In-rich InGaN QDs grown on n-GaN layers with different surface roughnesses. The forward operating voltage for the LEDs at an injection current of 20 mA was around 4.6 V. The higher operating voltage by 2 V compared to the InGaN MQW based LED is due to poor crystal quality of the p-GaN grown at 900 °C, which is 100 °C lower than the conventional growth temperature. Figure 4(a) shows the EL spectra of the LEDs measured at an input current of 20 mA. The dominant EL peak shifts to the longer wavelength side from 540 to 610 nm and broadens with increasing surface roughness of the underlying n-GaN layer from 21.3 to 36.2 Å. The broad EL spectrum for an LED with an rms roughness of 36.2 Å is composed of three elementary EL peaks at 570, 610, and 639 nm as plotted

by dashed lines (E1–E3). The EL peaks at 540 and 564 nm for the LEDs with rms roughnesses of 21.3 and 28.3 Å correspond to the QD ensembles producing P3 and P4 in the PL results, respectively. The EL peaks at E1 (570 nm) and E2 (610 nm) for the LED with an rms roughness of 36.2 Å correspond to the QD ensembles producing P4 and P5 in the PL results, respectively. Figures 3 and 4 show that the dominant EL peak position of the LEDs is redshifted compared with that of the PL spectra. This can be attributed to either a change in the size or composition of the In-rich InGaN QD ensembles by the high temperature overgrowth of the p-GaN layer for LED fabrication or a difference in the recombination process of PL and EL emissions. As shown in Fig. 4(b), the emission color of InGaN QD LEDs changes from green to red due to the increase of the surface roughness of the underlying n-GaN layer and the decrease of the compressive stress in the overgrown InGaN layers. These results indicate that the In-rich InGaN QDs formed by enhanced phase separation can be a promising light-emitting source for green gap spectral range LEDs.

In summary, green gap spectral range LEDs were fabricated using self-assembled InGaN QDs. The emitting wavelength was controlled by the surface roughness of the underlying n-GaN layer. By increasing the surface roughness of the n-GaN layer, the emission wavelength of the LEDs was controlled from the green to red spectral range. Structural and optical characterizations showed that phase separation in the InGaN layer is enhanced by increasing the surface roughness, which results in the formation of an InGaN QD ensemble with high In composition or large InGaN QDs. By controlling the surface roughness of the underlying n-GaN layer, In-rich InGaN QD LEDs, emitting the light from green to red spectral range, were demonstrated for use as a promising active source for green gap LEDs.

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