Investigation of the Indium Atom Interdiffusion on the Growth of the InGaN/GaN Heterostructure

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The influence of the growth procedure on the optical quality of InGaN grown on GaN has been investigated. The PL spectrum of the sample with a low-temperature-grown GaN cap layer or a graded-temperature-grown GaN cap layer has a shorter peak wavelength than that of the sample grown with a interruption procedure. The shift of the peak wavelength increases with the increase of the layer thickness of the low-temperature-grown GaN cap layer. This is because the defect contained in the low-temperature-grown GaN cap layer to induce the outdiffusion of In atoms during the temperature-ramped procedure. The narrower linewidths and high intensities of the InGaN after In outdiffusion may be due to the reduction of the strains, dislocations, or defects.

1. TEXT

The III-nitride based semiconductor with a wide and direct energy bandgap have emerged as the leading materials for short wavelength optoelectronic devices operating from the UV to blue wavelengths. The commercialization of bright blue InGaN/GaN double heterostructure (DH) based LEDs by Nakamura et al.¹⁾ has ignited considerable and rapidly increasing interest in the III-V nitride system. Epitaxy of InGaN has been performed by metal organic chemical vapor deposition (MOCVD)^{2),3)} and molecular beam epitaxy (MBE)⁴⁾. The single quantum well of the InGaN has been used as the active layer for high brightness blue, green, and yellow LEDs, and more recently as the InGaN multi-quantum-wells active layer in the blueviolet laser diodes (LDs) 5). Deposit the progress in the device development, only a few studies investigating the growth of InGaN by MOCVD have been reported. However, several problems are involved with the epitaxial deposition of indium contained in the InGaN layer. Understanding the growth of the ultra-thin InGaN/GaN quatum well by MOCVD is of extreme importance in improving the properties of these devices. That is because the deposition temperature required for the InGaN, which is determined by the high evaporation rate of indium, is far lower than that required for GaN. In addition, the decomposition rate of the group-V precursor ammonia drops from 60 % at 1000 °C down to about 7% at 900 °C, so a rather high deposition temperature is recommended. In this letter, we study the influence of the growth procedures with different temperature ramping steps on the characteristic of InGaN as the high deposition-temperature GaN grown above the InGaN layer. The 10K photoluminescence (PL) spectra and the room temperature raman spectra of the samples were analyzed.

The structure used in this study is shown in Fig.1. The epitaxial films were grown on the C-plane sapphire in a low

pressure MOCVD system. The substrate was pre-head in a H2 stream for 25 minutes at 1100 °C. After that, the temperature was down to 525 °C and a 10 nm thick GaN buffer layer was deposited. Then, the temperatute was raised to 1025 °C and a thick GaN layer was grown on the buffer layer. Before the 10 nm InGaN layer was grown at 750 °C, the growth interruption was used and the temperature was ramped down to 750 °C. Then, a 100 nm GaN cap layer was deposited. To study the interface characteristics of the GaN layer on a thin InGaN film, four samples with different temperature-ramped procedures were prepared. These procedures are included : (a) the growth temperature was directly ramped to 1025 °C and then a 100 nm high-temperature-grown (1025 °C) GaN cap layer was deposited, (b) a 10 nm GaN layer was deposited at 750 °C, after that, the temperature was directly ramped to 1025 °C and a 90 nm GaN cap layer was deposited at 1025 °C, (c) a 50 nm GaN cap layer was deposited during the growth temperature was graded ramped to 1025 °C, and then a 50 nm GaN was deposited at 1025 °C, and (d) a 50 nm GaN layer was deposited at 750 °C, after that, the temperature was directly ramped to 1025 °C and a 50 nm GaN cap layer was deposited. The different growth procedures are shown in Fig.2. After growth, 10 K photoluminescence (PL) measurement was performed. The 325 nm He-Cd laser was used as the exciting laser. The room temperature raman scattering is also measured in this study. The 441.6 nm He-Cd laser was used as the pumping laser. The power is 15 mW and the resolution is 3 cm⁻¹.

Fig 3. shows the PL spectra of the samples with different growth procedures. From Fig.3, it can be seen that the peak wavelengths of the 10 K PL spectra shift to the short wavelengths as the samples with a low-temperature-grown GaN cap layer. In addition, the shifts of the peak wavelengths are increased with the increase of the layer thickness of the low-temperature-grown GaN layer. This phenomenum may be due to the defects contained in the

low-temperatute-grown GaN layer to induce the interdiffusion of In atoms during the temperature ramping procedure. This is because the GaN layer grown at a low temperature has a large of defects within the layer to induce the diffusion of In atoms and the peak wavelengths of the PL spectra shift to short wavelengths.

From the PL spectra of samples (c) and (d), it can be seen that although the growth procedures are different for both samples, the wavelength shifts of the PL spectra for these samples have the same tendency and the peak intensities are stronger than the other two. In addition, the full width half maximum of the spectra for these two samples are smaller than that of sample (a), which indicates that the low-temperature-grown InGaN layer has become a high quality InGaN film with a low In mole fraction since the LT-GaN layer was deposited to induce the outdiffusion of In atoms into the LT-GaN layer. This phenomenum may be due to the reduction of the defects, strains, or the dislocation density. The details of such mechanism need more studies to identify.

The raman spectra of the samples are depicted in the Fig.4. As can be seen in Fig.4 , the peak intensity of E_2 is dramaticly decreased due to the different growth procedures. The sample A has the highest intensity among these four samples. This is because that the GaN cap layer of the sample A is totally grown at 1100 °C. The quality of the GaN film for the sample A is the best during these samples. In another word, the peak intensity of E₂ for the sample D with a 50 nm 750 °C grown GaN is the lowest. From Fig.4, another phenomenum needed to be considered is the competition of the A_1 (LO) and E_1 (LO) peaks. It can be seen that the sample grown without the low-temperature grown GaN, the intensity of the A₁(LO) is larger than that of E₁(LO). When the sample with a thicker lowtemperature-grown GaN, the intensity of the E1(LO) is larger than that of the $A_1(LO)$. This result may be due to the interdiffusion of In and Si atoms between InGaN and GaN. This is because the InGaN layer was doped with Si atoms to enhance the edge emission for these samples 6),7), the interdiffusion of Si atoms from InGaN to GaN is reasonable during the high temperature thermal treatment. In addition, the defects of the low-temperature-grown GaN to enhance the interdiffusion of the In atoms will also induce the phonon mode competition due to the non-perfect GaN crystal. So , from Fig.4, the sample grown with a lowtemperature-grown GaN, the intensity of the A1(LO) is smaller than that of the $E_1(LO)$.

Besides considered above, it can be seen that the peaks of $A_1(TO)$ and $E_1(TO)$ are appear in FIg.4. The existences of the $A_1(TO)$ and $E_1(TO)$ indicate that the c-axis of the GaN crystal is oblique. This result indicates that the InGaN layer grown on the thick GaN layer may be not uniform and induce non-uniform strian to result in the oblique of the c-axis for the GaN cap layer⁸. So, the peaks of $A_1(TO)$ and $E_1(TO)$ can be detected by the raman measurement. The details of the raman spectra need to be further studied.

In conclusion, we have investigate the influence of the growth procedure on the optical quality of InGaN grown on GaN. The PL spectra of the samples with a low-

temperature-grown GaN cap layer or a graded-temperaturegrown GaN cap layer will have a shorter peak wavelength than that of the sample grown by a abrupt temperatureramped rate. In addition, the shifts of the peak wavelengths are increased with the increase of the layer thickness of the low-temperature-grown GaN cap layer. This is because the defect contained in the low-temperature-grown GaN cap layer to induce the outdiffusion of In atoms during the temperature-ramped proceduce. The InGaN layer after diffusion has a low In mole fraction and the peak wavelength of the 10 K PL spectra shifts to the short wavelength than the sample without a LT-GaN layer. From this study, it also indicates that the InGaN layer with a graded-temperature-grown GaN or a 50 nm-thick LT-GaN cap layer will have a higher PL intensity. The narrower linewidths and high intensities of the InGaN after In outdiffusion may be due to the reduction of the strains, dislocations, or defects. For the device application, the LT-GaN cap layer or a garded-temperature-grown GaN cap layer can be used to improve the optical quality of the InGaN layer, but the In composition will be lower than than that without a such capped layer. From the raman scattering measurement, we find the In atoms interdiffusion to induce the phonon mode competition. In addition, the GaN cap layer grown on the InGaN will has a oblique c-axis crystal due to the non-uniform strains induced by the InGaN layer. For future device application, such a growth procedure will be easily used to improved the optical characteristics of the InGaN layer.

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2. FIGURES



Fig. 1. The schematic structure of the InGaN/GaN single quantum well grown on Al₂O₃.



Fig.2 The different growth procedures . (a) with a growth interruption, (b) with a 10 nm low-temperature-grown GaN, (c) with a 50 nm graded-temperature-grown GaN, (d) with a 50 nm low-temperature-grown GaN.



Fig.3 The 10 K PL spectra of the samples with the different growth procedures.



420 470 520 570 620 670 720 770 820 Raman Shift (cm⁻¹)

Fig.4 The room temperature raman spectra of the samples. The phonon modes from Al_2O_3 and GaN are also indicated.

3. REFERENCES

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