Journal of the Korean Vacuum Society
Vol.12, No.S1, 2003, pp.95~99

Effect of growth interruption on InN/GaN single quantum well structures

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Abstract

We successfully grew InN/GaN single quantum well structures by metal-organic chemical vapor deposition and confirmed their formation by optical and structural measurements. We speculate that relatively high growth temperature (730℃) of InN layer enhanced the formation of 2-dimensional quantum well structures, presumably due to high adatom mobility. As the growth interruption time increased, the PL emission efficiency from InN layer improved with peak position blue-shifted and the dislocation density decreased by one order of magnitude. The high resolution cross-sectional TEM images clearly showed that the InN layer thickness reduced from 2.5 nm (without GI) to about 1 nm (with 10 sec GI) and the InN/GaN interface became very flat with 10 sec GI. We suggest that decomposition and mass transport processes on InN during GI is responsible for these phenomena.

Keywords : InN/GaN single quantum well, growth interruption, metal-organic chemical vapor deposition

1. Introduction

Nitride semiconductors have received much attention for use in optoelectronic and electric devices [1-5]. However, InN has been much less studied than other nitride semiconductors. This is due to the fact that the thermal instability of InN as well as the large lattice and thermal mismatch make the growth of high quality InN films difficult. Because of thermal instability of InN, the general preparation of InN requires a low growth temperature. For common III-V nitride epitaxy techniques, such low growth temperature restricts an adatom migration distance. Therefore, many group III atoms will have less energy to travel long enough to locate their minimum energy sites on the surface. As a result, the InN film with high defect density will form and it is too difficult to obtain 2-dimensional (2D) InN epilayer. In spite of these problems, the growth of 2D InN epilayer on GaN substrate is inevitable for many potential applications such as high electron mobility transistors (HEMTs) and light emitting diodes (LEDs).

In this work, we report our experimental results on the effect of growth interruption (GI) on InN/GaN single quantum well (SQW) structures grown at relatively high temperatures (730℃) compared to generally reported InN growth temperatures [3-5]. From our experimental results, it was found that the introduction of GI in InN/GaN SQW structures improved the optical and structural properties in the quantum well (QW) layer. The detailed information on the effect of GI on InN/

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GaN SQW structures will be addressed.

2. Experiment

InN/GaN SQW structures were grown by metal-organic chemical vapor deposition (MOCVD). The details of our MOCVD system were described elsewhere [6]. The growth pressure was maintained at 300 Torr throughout the whole process. InN growth was performed during 90 sec on 2-μm-thick GaN epilayer grown on sapphire at 1080℃. Growth temperature of InN layer was 730℃. Then, 20-nm-thick GaN capping layer was grown with or without GI at the same temperature (730℃). The GI times varied from 0 to 30 sec. For effective NH3 cracking at low growth temperature [6] and reduction of native defect density [7], the ammonia was preheated before they entered into the reactor. The grown InN/GaN SQW structures were characterized by photoluminescence (PL), atomic force microscopy (AFM) and transmission electron microscopy (TEM). PL experiments were performed using a He-Cd laser (325 nm) or a Xe lamp as a pumping source. The prepared TEM specimens were examined by a JEOL JEM-3000F microscope operating at 300 kV with point-to-point resolution of 0.17 nm. In scanning TEM (STEM) mode, energy dispersive spectroscopy (EDS) analysis with an electron beam size of 1.5 nm was used for the compositional analysis of the QW layer.

3. Results and Discussion

From PL and TEM measurements, we found that the InN/GaN SQW structures were formed regardless of GI. We speculate that relatively high growth temperature of InN layer enhanced the formation of 2D QW structures, presumably due to high adatom mobility. From the low temperature (LT) PL measurements, we observed very strong PL peaks near 400 nm. To classify the origin of these PL peaks, we performed the PL measurements using a Xe lamp as a pumping source with various excitation wavelengths from 325 to 375 nm. From these measurements, we found that the PL peak near 400 nm was originated from the InN layer, not from the GaN layer. The possible reasons for the short emission wavelength from the InN/GaN SQW and the changes in emission wavelength in terms of quantum confinement effect was discussed in detail elsewhere [8].

As the GI time increased, the PL peak showed a blue shift, as shown in Fig.1. The integrated PL peak intensity increased and the full width at half maximum (FWHM) decreased by increasing GI time. These phenomena can be explained in terms of increased quantum confinement effect, which comes from the decrease in InN QW thickness as well as the improvement in InN/GaN interface quality, as confirmed by cross-sectional TEM (XTEM) analysis in Fig. 2.

From XTEM measurements, we observed that InN QW thickness varied from 2.5 nm to 1 nm by introducing GI time from 0 to 10 sec. The InN/GaN

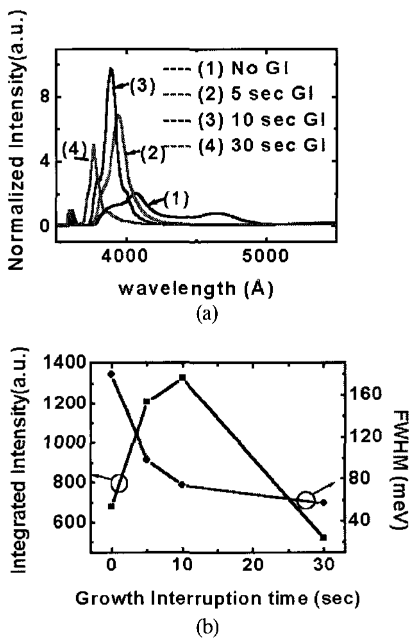
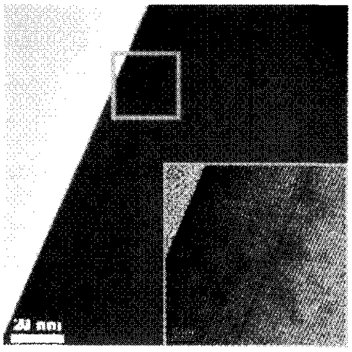
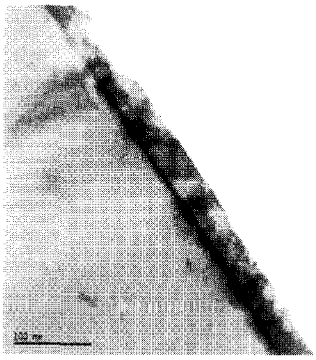


Fig. 1. LT (12K) PL spectra of (a) InN/GaN SQW structures and (b) integrated intensity and FWHM of PL peak as a function of growth interruption time.



(a)



(b)

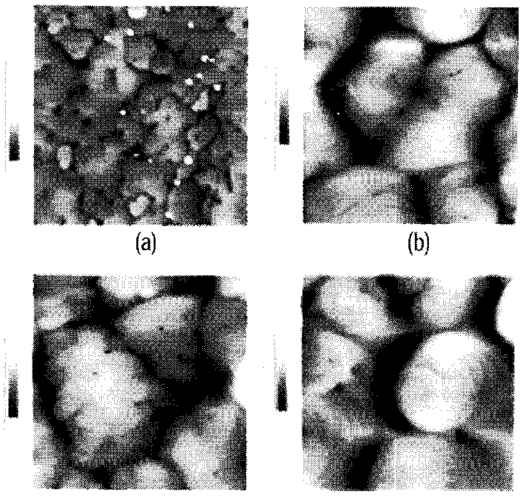
Fig. 2. Cross-sectional TEM images of InN/GaN SQW structures with (a) no GI and (b) 10 sec GI. The inset in (b) is the high-resolution TEM image.

interface became very flat with 10 sec GI, as shown in Fig. 2(b). Some contrast fluctuations in the QW region are visible in Fig. 2(a). It can be caused by InN thickness fluctuation when the incoming flux is high, compared to the lateral diffusional flux. The XTEM micrograph (Fig. 2(a)) also indicates that the crystalline quality of the GaN capping layer is relatively poor; the low-temperature capped GaN layer has thickness fluctuation as well as pits on the surface. On the other hand, the InN/GaN SQW sample with 10 sec GI showed the remarkably flat GaN capping layer with less defects as shown in Fig. 2(b). In high resolution TEM image, atomically flat InN/GaN hetero-interfaces are clearly observed.

To clarify the In concentration fluctuation in the InN QW region, we performed the EDS analysis in STEM mode. From parallel scan to QW layer, there were no considerable In and Ga compositional fluctuation in the QW layer. The clear In signal was observed from vertical scan of the InN QW region. Unfortunately, in this InN/GaN SQW structures, the QW layer thickness is very thin (~2.5 nm), similar to the electron probe beam diameter (~1.5 nm), so that we could not obtain the absolute In composition in the QW layer.

To quantify the defects in the InN/GaN SQWs at various GI times, the samples were examined by AFM. The sample without GI had dislocation density as high as 4×10^9 cm^{-2} as shown in Fig. 3(a). Considering the dislocation density of the GaN template layer (2.5×10^8 cm^{-2}), dislocation density is almost one order of magnitude higher. On the other hand, the samples with 10 sec GI had a dislocation density of 5×10^8 cm^{-2}, consistent with the XTEM image of Fig. 2(b).

From Matthews and Blakeslee's criterion [9], the critical thickness for misfit dislocation generation in InN/GaN structure is indeed less than 1 ML. And according to the in-situ analysis of Ng et al. [4], about 80% of the total strain in the epitaxial InN film grown



(a) (b)

(c) (d)

Fig. 3. 2 μm×2 μm AFM images of InN/GaN SQW structures with (a) no GI, (b) 5 sec GI, (c) 10 sec GI, (d) 30 sec GI. LT-grown GaN surface on GaN template typically shows a spiral growth mode like (b),(c) and (d).

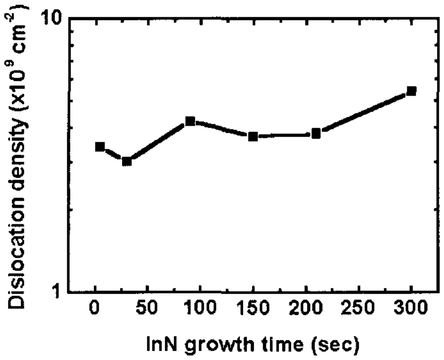


Fig. 4. Change of dislocation density in InN/GaN SQW structure with no GI as a function of InN growth time.

on GaN was initially relieved by defects (dislocations) within first 2 MLs. Further relaxation occurred at a very slow rate and contributed to island formation. Therefore, approximately 10 ML of InN formed in the sample without GI would be quite exceeding the critical thickness and it is not surprising to see the increase in dislocation density through dislocation generation. As we can see in Fig. 4, the dislocation density already reaches up to $3\sim4\times10^9$ cm^{-2} at 5sec InN growth time (~1 ML). This means that the dislocation already formed in the initial stage of InN growth to eliminate the misfit strain generated from very large lattice mismatch between InN and GaN.

For the InN/GaN SQW structure grown without GI, we believe that the growth rate of InN was relatively high, compared to lateral diffusion, so that the atoms had little chances to move to their minimum energy sites. Thus, at the initial stage of the InN growth, most of the misfit strain would be relieved by generation of dislocation and the remaining strain would be diminished by island formation, as we can see in Fig. 5(a). Therefore, as-grown InN layer on GaN would have a rough surface with many islands and structural defects. By introducing GI, however, atoms would have more chances for migration to find their minimum energy sites and defects in InN layer would be annihilated so that the structural quality of InN layer improved significantly like Fig. 5 (b). There would be severe decomposition of InN

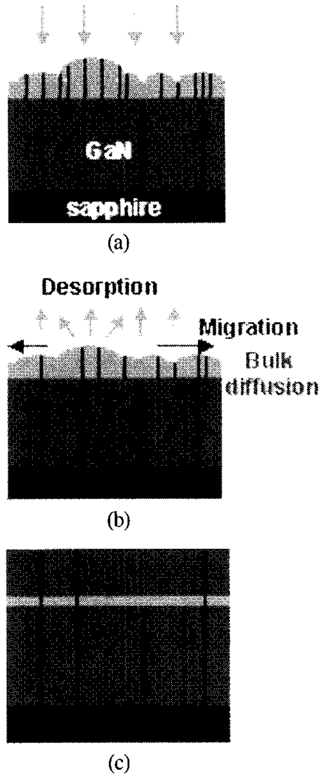


Fig. 5. Schematic diagram of effect of growth interruption in InN/GaN SQW structures: (a) during InN deposition (b) during GI (c) after GI

during GI due to relatively high growth temperature so that InN QW thickness decreases as GI time increases. Furthermore, because of the more drastic decomposition of the InN at regions of swelling surface during GI, the InN surface would be more flattened after GI. Therefore, after GI, the InN layer would have a smooth surface with low structural defect density and subsequently overgrown LT-capped GaN layer would have a low defect density as good as that of underlying GaN template as shown in Fig. 5(c).

4. Conclusions

We found that the InN/GaN SQW structures were formed regardless of GI. We speculate that relatively high growth temperature for InN layer enhanced the formation of 2-dimensional quantum well structures, presumably due to high adatom mobility. By increasing GI, the PL peak position blue-shifted with increased PL emission efficiency. The structural quality of InN/GaN SQW improved significantly. We suggest that the decomposition and mass transport process on InN during GI is responsible for these phenomena.

Acknowledgments

This work was supported by Samsung Advanced Institute of Technology, by the Ministry of Science and Technology of Korea through the National Research Laboratory Program and by the Ministry of Education through the BK21 Program.

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