Influence of AlGaN Buffer Growth Temperature on GaN Epilayer based on Si(111) Substrate

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Abstract. This paper investigated the influence of AlGaN buffer growth temperature on strain status and crystal quality of the GaN film on Si (111) substrates by metal organic chemical vapor deposition. It was demonstrated by the optical microscopy that AlGaN buffer growth temperature had a remarkable effect on compensating tensile stress in top GaN layer and preventing the formation of cracks. X-ray diffraction and atomic force microscopy analysis showed crystal quality and surface morphology of the GaN epilayer could be improved through increasing AlGaN buffer growth temperature. 1 μm crack-free GaN epilayer on Si (111) substrates was obtained with graded AlGaN buffer layer at optimized temperature of 1050℃. Transmission electron microscopy analysis revealed that a significant reduction in threading dislocations was achieved in GaN epilayer.

1. Introduction
Gallium nitride (GaN) compound semiconductor material system has been extensively of great interest for the large band gap, high breakdown electric strength, high electron saturated drift velocity and good thermal stability. Furthermore, the particular spontaneous and piezoelectric polarization effects lead to high concentration of two-dimensional electron gas (2DEG) near the interface of AlGaN/GaN hetero-structure. Consequently, GaN material has a large potential application in the areas of high power and high frequency electronic devices, light emitting diodes (LEDs) and laser diodes [1-5]. Generally GaN is grown on sapphire, SiC and Si substrates. Sapphire substrates limit the improvement of the power density of the device because of the bad thermal conduction. Meanwhile, the high cost and small size of SiC substrates badly hinder the application of GaN material. Compared with sapphire and SiC, Si is the best alternative substrate for its low cost, good thermal conductivity and ability to be integrated with the mature Si-based processing techniques. However, the large lattice mismatch (17%) and coefficients of thermal expansion (CTE) mismatch (56%) between GaN epilayer and Si would induce large residual tensile stress in the GaN based epilayers [6,7], which lead to the formation of cracks and high defect density that seriously degrade the performance of GaN based devices on Si substrate. Thus the design of suitable buffer layers towards the reduction of residual stress is crucial. Several approaches, such as introduction of compressively strained layers or a bridging interlayer with similar CTE, epitaxial lateral overgrowth using patterned Si substrates, have been developed to
compensate for the large thermal tensile stress and prevent the formation of cracks [8-14]. Among these high temperature (HT) AlN buffer is a simple and effective way to relieve tensile stress and further prevent cracks in GaN epilayer. However extra threading dislocations (TD) will be introduced into top GaN due to large cryatal lattice mismatch between AlN and GaN [15,16]. In our study, we grow an AlGaN buffer layer on HT AlN buffer prior to GaN growth. Compared with a single AlN buffer, the AlN/AlGaN multibuffer facilitates the compensation for tensile stress in GaN film, as well as reduce the TD density, owing to the crystal lattice of AlGaN between those of AlN and GaN.

In this work, we studied the effect of AlGaN buffer growth temperature on the surface morphology, strain status and crystal quality of GaN epilayer on Si(111) substrates by comparing five samples with AlGaN buffer of different growth temperature. 1μm crack-free GaN epitaxy with graded AlGaN buffer at optimized temperature was presented. It was observed by transmission electron microscopy (TEM) to investigate the TD evolution from through AlGaN buffer layer.

2. Experimental

In this work, the growth of samples was performed in a horizontal metal organic chemical vapor deposition (MOCVD) reactor on Si(111) substrates. Prior to loading in the growth chamber, the Si substrates were etched in boiling sulphuric acid, HCl:H2O2:H2O(1:1:5) solution, NH4OH:H2O2:H2O(1:1:5) solution and diluted HF solution in turn, which resulted in oxide-free hydrogen-terminated Si(111) surface. An in situ hydrogen exposure at 1100 ℃ was performed immediately after the substrates were loaded. Trimethylgallium(TMG), trimethylaluminum(TMA) and ammonia(NH3) were used as Ga, Al and N precursors, respectively, while N2 and H2 as carrier gases. The entire GaN epitaxial layers containing of a HT AlN buffer and an AlGaN buffer followed by 1μm GaN film was shown in Fig. 1. Five samples with different AlGaN growth temperature were investigated, labeled by samples A, B, C, D and E, respectively, as listed in Tab.1. For samples A, B, C and D, the Al composition of AlGaN buffer was fixed at 30% uniformly, with AlGaN growth temperature varied from 900, 1000, 1050, to 1080 ℃. For sample E, the AlGaN buffer growth temperature was 1050 ℃ while a graded AlGaN buffer layer instead of Al-composition-constant ones with the Al composition modulated gradually in the range of 30% to zero from the bottom to the top. Before AlN growth, the Si(111) substrates were exposed to TMA flow for several seconds without any ammonia, for the purpose of depositing an ultra Al layer to prevent the formation of a silicon nitride phase on the Si surfaces. The HT AlN buffer layer, and GaN film were grown at 1050 ℃ and 1060 ℃, respectively. The pressure was set to 100Tor for all deposition steps.

Optical microscopy (OM) and atomic force microscopy (AFM) were used to evaluate the crack density and surface morphology of GaN film. X-ray diffraction (XRD) was performed to measure the full width at half maximum (FWHM) of GaN(0002) rocking curve peaks to characterize the crystal quality. TEM was carried out to survey the TDs evolution with graded AlGaN buffer at optimized temperature.

3. Results and discussion

Crack density and surface morphology of the samples were investigated by OM. No white pits appeared to naked eyes on the surface of samples A, B, C and E. However some were presented in sample D. Under OM observation we found some circular defects on the surface of sample D (shown in Fig. 2a) while mirror-like and specular surfaces were shown in other four samples (OM image of sample E was shown in Fig. 2b). Dadgar reported the circular defects were caused by reaction of Ga and Si atoms outdiffusing from substrate during the high temperature growth process, and called it melt-back etching reaction [17]. The melt-back etching reaction of Ga and Si atoms is a well known problem of GaN epitaxy on Si, which resulted in circular defects and was severely destructive to the crystal quality of the GaN film [18]. The absent of circular defects in samples A, B, C and E proved the growth temperature (below 1050 ℃) of AlGaN buffer were proper to avoid melt-back etching reaction while the AlGaN growth temperature of sample D was so high that led to Si atoms diffusing out. A.M.Sanchez interpreted the circular defects origination by step theory [19].
The maximum crack-free ranges of the five samples were shown in Tab. 1. Samples A, B, D exhibited cracks across the entire wafer surface in three sets of parallel arrayed at 120° to one another, with the crack-free ranges of 9mm×10mm, 13mm×14mm, 11mm×11mm, respectively. In sample C the cracks were only found near the edges of the wafer while sample E revealed a crack-free surface on the overall wafer as shown in Fig. 2b. The results indicated AlGaN growth temperature influenced the crack density obviously and the optimized temperature was existed. It is known that the AlN interlayer will impose a compressive stress to the GaN epilayer which can partially offset the thermal tensile stress during the cooling down process. However, the GaN layer grown on the AlN buffer tends to relax due to the 2.4% lattice mismatch between GaN and AlN. The additional introduction of an AlGaN buffer layer with proper growth temperature, especially the grated AlGaN buffer, between GaN and AlN could lead to a soft transition in the lattice constants. Therefore, the compressive lattice mismatch stress may be more effectively maintained to compensate for the tensile stress induced by CTE mismatch. If the AlGaN growth temperature was not high enough the crystal quality was inferior, thus the compensation effect was limited. But if the growth temperature was too high, the thermal tensile stress became larger, and cracks emerged again.

The surface morphology of sample was characterized by AFM. The root mean square roughness (RMS) of sample was considerably decreased with AlGaN buffer growth temperature increasing. Meanwhile it was clearly seen the RMS of sample with graded AlGaN (sample E) was smaller than that of sample with Al-composition-constant ones (sample C) at the same growth temperature, due to the threading dislocations decreased by the employment of graded AlGaN. The results revealed that surface morphology of GaN film could be improved by increasing AlGaN growth temperature. Especially, it proved graded AlGaN buffer was more appropriate to perfect the surface morphology than the Al-composition-constant ones at the same growth temperature. The AFM image with 2μm×2μm scan scope of sample E was shown in fig. 3, which exhibited smooth surface with a RMS of 1.24nm.

The X-ray measurements for samples were crried out with symmetric GaN(0002) reflection using ω scan. GaN(0002) XRD FWHMs of rocking curve peaks were used to investigate the influence of AlGaN growth temperature on the crystal quality of GaN film. Results were shown in Fig. 4. It was clearly seen that FWHMs of samples decreased with AlGaN growth temperature increasing. It’s suggested the AlGaN buffer crystal quality was improved with the growth temperature raising. Better buffer crystal quality led to better GaN epilayer crystal quality. Thus FWHM decreased with AlGaN growth temperature increasing. On the other hand, it was found FWHM of sample with graded AlGaN (sample E) was smaller than that of sample with Al-composition-constant AlGaN buffer (sample C) at the same growth temperature. Structure with graded AlGaN seemed more helpful to improve the crystal quality than that with the Al-composition-constant ones. These XRD results suggested that a graded AlGaN greatly improves the in-plane crystalline quality of GaN. The reason was that the gradual decrement of Al content resulted in a modest change in the lattice constants within AlGaN buffer layer and it could be expected to maintain a pseudo-2D growth mode for the successive GaN layer. Superior crystal quality of GaN film was presented due to the pseudo-2D growth mode. In another way, the decrease in FWHM could be attributed to a reduction in the dislocation density by the graded AlGaN buffer. Detailed discussions would be supplied by TEM analysis in the following.

In order to analyze the mechanism to achieve high crystal quality GaN on Si(111), cross-sectional TEM was performed. The electron beam is along the [11 00] zone axis so that all types of TDs can be revealed as shown in Fig. 5. A large number of TDs were clearly seen originating from the AlGaN/AlN interface due to the mismatch between AlGaN and AlN layers[20] and thread through the whole graded AlGaN buffer. Significant dislocation reduction took place at the AlGaN/GaN interface while few threading dislocations generated at the interface. Obviously the GaN layer owned lower TD density than the AlGaN buffer. Thus, we could declare the AlGaN graded buffer layer acted as a dislocation filter. Another dislocation reduction took place in the first 500nm GaN above the AlGaN buffer, where a large number of the residual TDs bent and interacted as shown in Fig. 5. It is clearly seen that TDs in the AlN buffer and the AlGaN buffer layer were normally perpendicular to the
growth surface. Whereas TDs in GaN bent and deviated away from the growth direction in the region next to the AlGaN buffer layer. These inclined TDs interacted with those possessing opposite Burger vector and finally annihilated, resulted in the reduction of most TDs in GaN epilayer and the compressive strain relaxation[21]. Relaxation of compressive stress was produced by such bending due to the formation of in-plane misfit segments [22]. It was reported that TD inclination assisted stress relaxation in compressively strained structures[23,24]. By annihilation of bent dislocations, the TD density decreased during the growth of the GaN layer associated with compressive stress relaxation. Vice versa, the bend of dislocation symbolized the intense compressive stress exerted by the AlN/AlGaN multilayer.

4. Conclusion
This paper was concentrated on the effect of AlGaN growth temperature on the crack density and crystal quality of GaN epilayer on Si(111) substrates. We found an optimized temperature of 1050℃ was existed to maximize the compensation effect caused by the compressive stress from AlN buffer. Crystal quality seemed improved with AlGaN growth temperature increasing. 1μm crack-free GaN was obtained using graded AlGaN buffer at the optimized growth temperature with GaN(0002) XRD FWHM of 603.6arcsec. It was confirmed by TEM analysis that graded AlGaN with optimized temperature made a significant reduction of threading dislocations in GaN epilayer, thus improved the crystal quality.

Acknowledgements:
This work has been supported by the Knowledge Innovation Engineering of Chinese Academy of Sciences (No. YYYJ-0701-02); the National Nature Sciences Foundation of China (No. 60890193, 60906006); the State Key Development Program for Basic Research of China (No. 2006CB604905, 2010CB327503), and the Knowledge Innovation Program of the Chinese Academy of Sciences (No. ISCAS2008T01, ISCAS2009L01, ISCAS2009L02).
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Figure Captions:

Fig. 1 Schematic diagram of sample structure

Fig. 2 OM images of sample D (a) and sample E (b)

Fig. 3 2μm × 2μm scope AFM image of sample E

Fig. 4 GaN(0002) XRD FWHMs of samples vs AlGaN growth temperature

Fig. 5 Cross-sectional TEM image of sample E where the electron beam is along the [1 1 0 0] zone axis.

Table captions:

Tab.1 detailed description of the five samples
Fig. 1
Fig. 2
Fig. 3
Fig. 4
Fig. 5
| Sample                      | A      | B      | C      | D      | E      |
|-----------------------------|--------|--------|--------|--------|--------|
| AlGaN temperature (°C)      | 900    | 1000   | 1050   | 1080   | 1050   |
| Crack-free range (mm²)      | 9×10   | 13×15  | Except | 11×11  | All the area |
| XRD GaN(0002)               | 674.2  | 637.9  | 610.5  | 594.6  | 603.6  |
| FWHM(arcsec)               |        |        |        |        |        |