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Epitaxial Growth of GaN by Gas Source Molecular Beam Epitaxy Using NH_3

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Epitaxial growth processes of GaN on gamma-(γ)Al_2O_3/Si(100) and γ-Al_2O_3/Si(111) substrates have been investigated by gas source molecular beam epitaxy using NH_3 as a nitrogen source with ***in-situ*** reflection high-energy electron diffraction (RHEED) observation. The γ-Al_2O_3 layer was epitaxially grown on Si substrates and used as an intermediate layer. The thickness of γ-Al_2O_3 is about 0.6 μm and the surface morphology is almost flat. It is found that the nitridation of Si surface is effectively suppressed with γ-Al_2O_3 layer. The GaN under cubic phase was epitaxially re-grown on γ-Al_2O_3/Si(100) substrate and hexagonal GaN with flat surface on γ-Al_2O_3/Si(111).

Key wards: GaN, NH_3, GS-MBE, RHEED

1. INTRODUCTION

The GaN and compounds of AlGaN are only promising materials for high-power devices and opto-electronic devices in the ultra violet-wavelength region [1-3]. Many GaN-based opto-electronic devices are demonstrated on sapphire and SiC substrates [4-6]. On the other hand, the GaN-based devices on silicon (Si) substrates have not been performed regardless of many advantages of Si such as high quality, low cost, controllability of the conductivity and effectiveness for the application combined with Si-ULSI technology [7,8]. It is hard to obtain high quality GaN epilayer on Si because it is easy to occur the interaction at the hetero-interface between GaN layer and Si substrate [9]. Therefore, it has been expected to grow a device-quality GaN epilayer on Si using an appropriate buffer layer [10-19].

In the present work, we have investigated the growth behavior of GaN epilayer on Si(111) and Si(100) substrates covered with 0.6 μm thick γ-Al2O3 epitaxial layer as an intermediate layer by NH_3 gas source molecular beam epitaxy (GS-MBE) with ***in-situ*** reflection high-energy electron diffraction (RHEED) system.

2. EXPERIMENT

Figure 1 shows a schematic diagram of a growth apparatus equipment with an ***in-situ*** RHEED system. A growth chamber was evacuated by twin trubo molecular vacuum pumps mounted in two lines individually, and the base pressure was of 1×10^{-10} Torr. γ-Al_2O_3 / Si(100) and γ-Al_2O_3 / Si(111) wafers were used as substrates. It was reported that a single γ-Al_2O_3 layers were epitaxially grown on Si(100) and Si(111) with mirror-like surfaces by low-pressure chemical vapor deposition [20-22] and MBE[23] methods, respectively. There were chemically cleaned with etching by dipping in a solution of before $HF:H_2O$ = 1:50 before being set in the preparation chamber. The substrate heated by directly passing a dc electric current through molybdenum electrodes. Prior to keeping a growth temperature of 600 to 870 ℃, the substrate were thermally cleaned at 900 ℃.

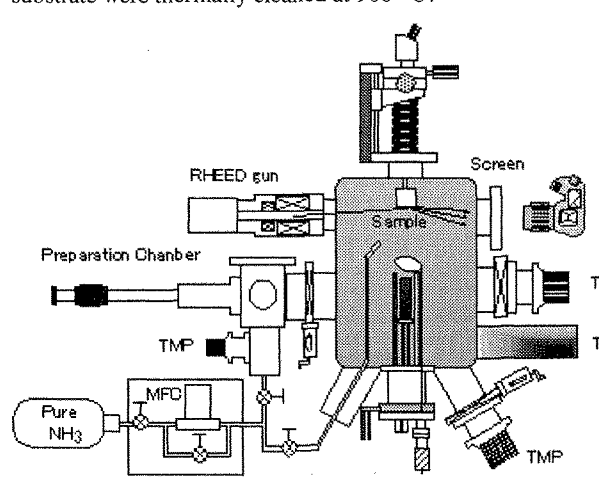

Fig. 1 Schematic diagram of growth apparatus evacuated by twin trubo molecular pumps with a RHEED system

The substrate temperature was measured by an optical pyrometer. NH_3 gas was introduced onto the substrate surface through a delivery stainless tube. The pressure of NH_3 gas during GaN growth was preciously controlled by a mass-flow controller in the range of 2×10^{-6} to 2×10^{-4} Torr . The substrate surface and film growth processes in the initial stage were observed ***in-situ*** by the RHEED system operated at an acceleration voltage 10 kV. The glancing angle of incident electron beam to the substrate surface was about 1.0° . The growth sequence was observed by digital still camera with mega-pixel CCD detector.

3. RESULTS AND DISCUSSIONS

3.1 Surface stability of γ-Al_2O_3/Si(100)

Figure 2(a) shows a RHEED pattern taken from a γ-Al_2O_3/Si(100) surface after thermal cleaning at a substrate temperature of 900 ℃ . In this figure, 1/3-order fractional super diffraction spots (indicated by downward allows) are observed between fundamental diffraction rods which two-dimensional Miller indices are (00) and (11). It is found that epitaxial γ-Al_2O_3 layer deposited on Si(100) has reconstructed structure having an three-hold periodicity in the [001] and $[00\bar{1}]$ directions. Figure 2(b) shows the RHEED pattern from the substrate surface after starting NH_3 gas irradiation.

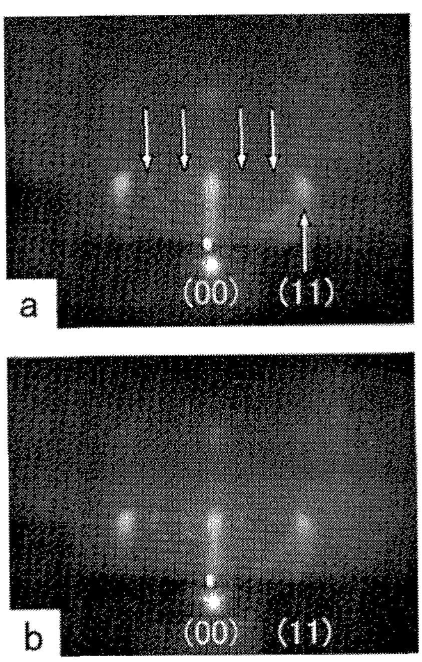

Fig. 2 RHEED patterns taken from γ-Al_2O_3 /Si(100) substrate surface after (a) thermal cleaning at 900 ℃ and (b) NH_3 gas irradiation. Incident electron beam is parallel to $[001]_{Si}$ direction.

It is noticed that there is no change in these RHEED patterns of fig. 2(a) and (b). This result strongly suggests that the surface nitridation of Si substrate is successfully suppressed by using the γ-Al_2O_3 epilayer as the intermediate layer.

3.2 GaN on γ-Al_2O_3/Si(100)

In order to investigate the growth process of GaN epitaxial growth layer and epitaxial relationship between the epilayer and substrate, the GaN epilayer is grown on γ-Al_2O_3 /Si(100) substrate directly. Fig. 3 shows RHEED patterns taken from GaN epilayer deposited for 30 min at the growth temperature of 870 ℃. The incident electron beam is parallel to fig. 3(a) [001] and (b) [011] directions. The sample was always exposed to NH_3 gas irradiation through the growth processes.

The distance ratio estimated by d_{11}/d_{10} nearly equals to square root of two, where d_{11} and d_{10} are the spacing between (00) and (11) rods showing in fig. 3a and (00) and (10) rods in fig. 3(b), respectively. This is meaning that the crystal structure of the GaN epilayser grown on γ-Al_2O_3/Si(100) has the cubic one. It is found that extra diffraction spots along the Laue circles in addition to the fundamental diffraction spots of a plane cubic structure. It is strongly suggested that the GaN epilayer is composed of many crystalline domains having tilted C-axis.

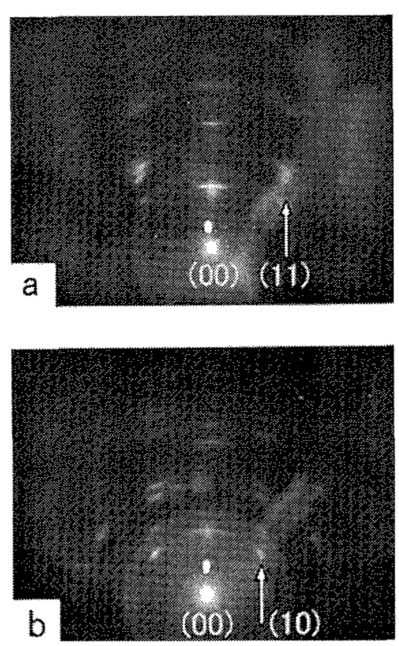

Fig. 3 RHEED patterns taken from GaN epilayer grown for 30 min on the γ-Al_2O_3 /Si(100) substrate at 870℃, where the temperature of Ga cell was 800℃ and NH_3 flow rate was 5 sccm. The incident electron beam is parallel to (a) [001] and (b) [011] directions.

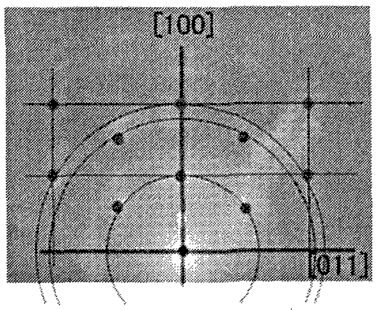

Fig. 4 Schematic diagram for explaining the RHEED pattern of fig. 3b. Solid circles are show the fundamental diffraction spots for a plane cubic GaN. Big circles indicate Laue zones.

3.3 GaN on γ-Al_2O_3/Si(111)

In order to investigate the growth process of GaN epilayer and the epitaxial relationship between the epilayer and substrate, the GaN epilayer is directly grown on γ-Al_2O_3 /Si(111). Fig. 5 shows ***in-situ*** RHEED patterns taken from (a) γ-Al_2O_3 /Si(111) surface after thermally cleaning at a substrate temperature of 900℃, (b), (c) and (d) GaN epilayers growing for 60min at a growth temperature of 800℃. The incident electron beam is parallel to fig. (a), (b) and (c) $[110]_{Si}$ and (d) $[112]_{Si}$ direction, respectively. It is found that RHEED pattern changes to 3-dimensional diffraction pattern as showing in fig. 5(b). This means that the islanding growth of the hexagonal GaN occurred at the initial growth step. These diffraction spots become weak and sharp two-dimensional (2D) diffraction streaks appear as proceeding of the growth of GaN layer in figs. 5(c) and 5(d). It is found that the growth mode of GaN epilayer changes form the islanding growth one to a 2D growth one. It is noticed that the diffraction streaks on the first order Laue zone indicating L_1 are clearly observed in fig. 5(d) in addition to the ones on the zeroth order Laue zone (L_0). It is considered that the growing surface of GaN layer has a extreme flat surface regardless of the direct growth method.

The lattice spacing of epitaxial layer parallel to the growing surface agrees with the one of GaN, which is determined by the distance between (00) and (10) diffraction rods in fig. 5(c) and (00) and (11) in fig. 5(d). Therefore, it is found that the GaN is epitaxialy grown on γ-Al_2O_3/Si(100) in the single crystal phase having flat surface under the epitaxial relationship with $[11\underline{2}0]_{GaN}$ // $[1\underline{1}0]_{\gamma\text{-}Al2O3/Si}$. Further discussions on the growth mechanism will be made using crystalline nucleation theories.

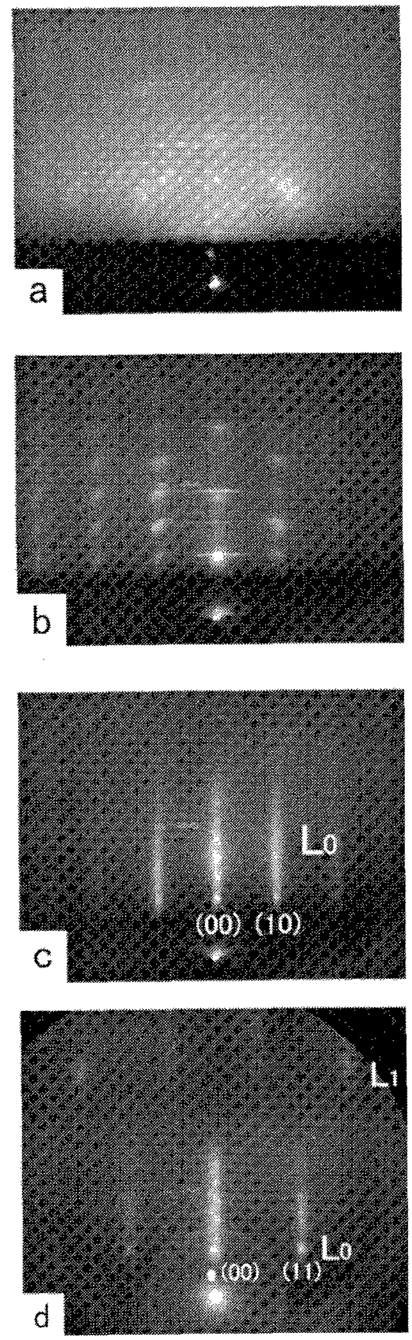

Fig. 5 RHEED patterns taken from γ-Al_2O_3 /Si(100) substrate surface after (a) thermal cleaning at a temperature of 900℃ and (b) growth time is 10 min (c) growth time is 60min, (d) 60 min. Incident electron beam is parallel to $[1\underline{1}0]_{Si}$ direction for (a), (b) and (c) and $[11\underline{2}]_{Si}$ for (d). Growth temperature is 800 ℃

4 CONCLUSIONS

Surface stability of γ-Al_2O_3/Si substrate and growth processes of GaN on γ-Al_2O_3/Si(100) and γ-Al_2O_3/Si(111) substrates using NH_3 gas source MBE method. The substrate surface of Si covered with γ-Al_2O_3 epilayer is very stable for irradiation of NH_3 gas at the growth temperature of 900℃. The single phase-crystalline of cubic GaN is grown on γ-Al_2O_3/Si(100). The single hexagonal GaN epilayer with smooth surface is successfully grown on γ-Al_2O_3/Si(111).

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