

Two Dimensional Growth of GaN on Various Substrates by Gas Source Molecular Beam Epitaxy Using RF-Radical Nitrogen Source

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Two dimensional (2D) layer by layer growth conditions for GaN epitaxial layers by gas source molecular beam epitaxy using a 13.56 MHz RF-radical nitrogen source were systematically investigated for the first time. It was clarified that the lower growth rate ($R < 0.1 \mu\text{m/h}$) enhanced the 2D growth. Furthermore, the introduction of annealed thin GaN buffer layer (60 Å) drastically enhanced the 2D growth for the layers grown on (0001) Al_2O_3 , (001) MgO and (001) GaAs substrates.

1. INTRODUCTION

Since the p-n junction blue light emitting diodes were realized by metalorganic chemical vapor deposition (MOCVD)^{[1],[2]}, III-V nitride semiconductors have been intensively investigated as attractive materials for short wavelength light emitting, high-temperature, and high-power devices.

In the late 1960s, the syntheses of nitride semiconductors were started, followed by many efforts made for obtaining high quality and p-type conductive nitrides, here using MOCVD, halide vapor phase deposition (HVPE), molecular beam epitaxy (MBE), which history was systematically reviewed by Pankove^[3], Davis^[4], and Strite^{[5],[6]}.

The MBE technology using reactive nitrogen (N_2) sources are directed close attentions as one of the superior growth techniques for nitrides, because of the reduced growth temperature, the possibility of as-grown p-type layers, and the high compositional controllability in monolayer thickness. The low temperature growth can bring about stoichiometric epitaxial layer growth at relatively low nitrogen supply ratio. Several types of the reactive nitrogen sources, such as the electron cyclotron resonance (ECR) microwave plasma source^{[7]-[9]}, the low-energy ion source^{[10],[11]}, and the RF-radical plasma source^{[12],[13]} have been utilized thus far and p-type GaN layers were obtained using the former two nitrogen sources. On the other hand, by use of the RF-radical sources, growing high quality GaN films was expected due to their lower ionic species generation rate in reactive nitrogen beams as compared to the ECR or the

ion sources^[12], which feature of RF radical source may result in the elimination of the ion bombardment damages from the epitaxial layer. However, there were only a few reports about GaN grown by the RF-radical nitrogen source and the properties of GaN layers were also not clarified.

In this work, we report the initial studies on the GaN growth by MBE using the RF-radical nitrogen source (RF-radical MBE) on various kinds of substrates.

2. EXPERIMENTS

In this investigation, (0001) Al_2O_3 , (001) MgO and (001) GaAs substrates were used. After the thermal cleaning at 600-610 °C, the substrates were exposed under RF-radical nitrogen beam for 5 minutes and then GaN layer was grown successively. In the growth, the substrate temperature, the nitrogen flow rate and RF input power were set to be 620 °C, 4.0 sccm, and 400 W, respectively. The growth rate R was changed as 0.15, 0.10, 0.075, and 0.05 $\mu\text{m/h}$. As we reported, the 5 minutes initial substrate nitridation brought about the growth of hexagonal GaN on (001) GaAs substrates^[13]. For all cases in the above, therefore, hexagonal GaNs were grown.

3. RESULTS AND DISCUSSION

Figure 1 shows the growth mode transition thickness for the GaNs on (0001) Al_2O_3 substrates, that is from island (3D) growth to layer by layer (two dimensional (2D)) growth mode, as a function of growth rate R . For

the thickness above the transition thickness, the 2D growth was proceeded. Here the growth mode transition was confirmed from the in-situ observation of reflection high energy electron diffraction (RHEED) patterns, that is, by the change point from the spotty pattern to the streak one. When the growth rate was 0.15 $\mu\text{m/h}$, the growth mode did not transfer to 2D growth mode even after 0.3 μm growth. On the other hand, for the growth rate of 0.1, 0.075 and 0.05 $\mu\text{m/h}$, the RHEED patterns changed to streak one after growing as 0.2, 0.06 and 0.04 μm in thickness (see open circles in Fig. 1). These results suggest that the lower growth rate and/or the higher V/III ratio enhanced the 2D growth of GaN.

The GaN buffer layer was introduced along the growth temperature time schedule illustrated in Fig. 2, where the RF input power was increased to 450 W and the growth rate was 0.1 $\mu\text{m/h}$. After growing 60 \AA GaN buffer layer, the Ga and nitrogen shutters were closed for annealing at 680 $^{\circ}\text{C}$ about 2 minutes. Then the substrate temperature was cooled down to 620 $^{\circ}\text{C}$ to start the GaN growth at the same conditions. In this case, the growth mode transition thickness drastically reduced to 160 \AA (see a closed circle in Fig. 1). This drastic enhancement of 2D growth was not observed for the following two cases; i.e. when the RF input power was 450 W without annealed GaN buffer layer and 400 W with annealed GaN buffer layer. From these results, we can consider that the use of an annealed thin GaN buffer layer with enough reactive nitrogen supply can drastically enhance the lateral 2D growth of GaN.

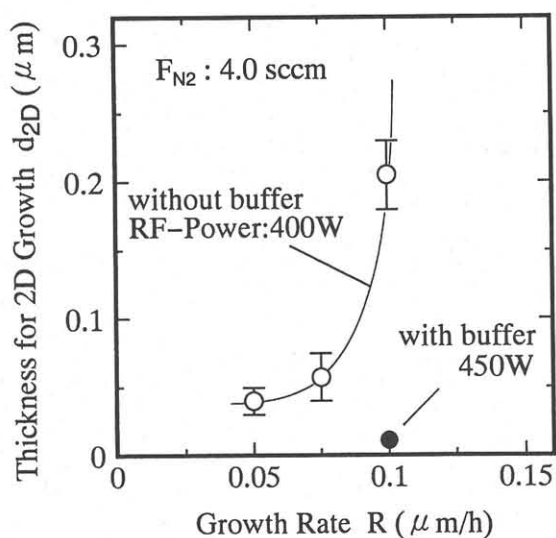


Fig. 1. Growth mode transition thickness (around this thickness, growth mode changes from island growth to layer by layer growth) as a function of growth rate.

Similar results were observed on (001) MgO and (001) GaAs substrates.

Fig. 3 shows the RHEED patterns of GaN layers after growing as 0.3 μm on (0001) Al_2O_3 , (001) MgO and (001) GaAs, respectively. These layers were grown with GaN buffer layers and 450 W RF input power. For all the samples, clear streak RHEED patterns were observed. While the slight difference observed in the sharpness of the streak rods suggested that the crystal structural uniformity of GaN was influenced by the material difference of substrates and superior as an order of GaN on (0001) Al_2O_3 , (001) MgO, and (001) GaAs.

The photographs of surface morphologies of these GaN layers observed by Nomarski microscope are shown in Fig. 4. The layer grown on (0001) Al_2O_3 had a mirror like featureless smooth surface (Fig. 4 (a)). The morphology of the layer grown on (001) MgO was also mirror like while having slightly rough texture (Fig. 4 (b)). For the whole surface of GaN on (001) GaAs substrates, however a rough texture and hexagonal cracks were observed (Fig. 4 (c)). These results were consistent with RHEED pattern observation results above mentioned.

The structure of these layers were determined by single crystal X-ray diffract meter using $\text{CuK}\alpha$ radiation. The diffraction peaks from the all GaN layers were observed between 34.55 and 34.78 degrees, corresponding to the (0002) diffraction of hexagonal GaN ($c=5.166 \text{\AA}$). That is, the c-axis of GaN layers was aligned in perpendicular to the substrate surface.

Electrical properties of the GaN layers grown on (0001) Al_2O_3 substrates were measured at r.t. by the Van der Pauw method using Al ohmic contacts. The high n-type carrier concentration over 10^{19} cm^{-3} was observed.

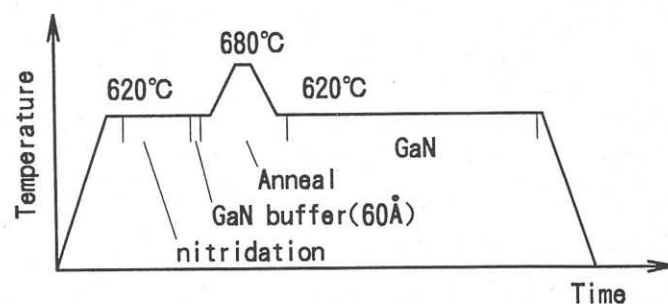


Fig. 2. Time schedule of GaN growth with thin GaN buffer layer.

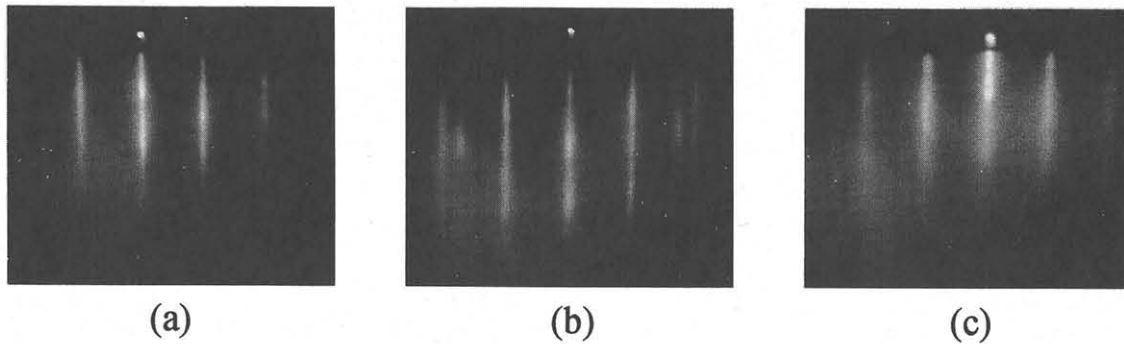


Fig. 3. RHEED patterns of GaN grown on the various substrates, (a): (0001) Al_2O_3 , (b): (001) MgO, and (c): (001) GaAs.

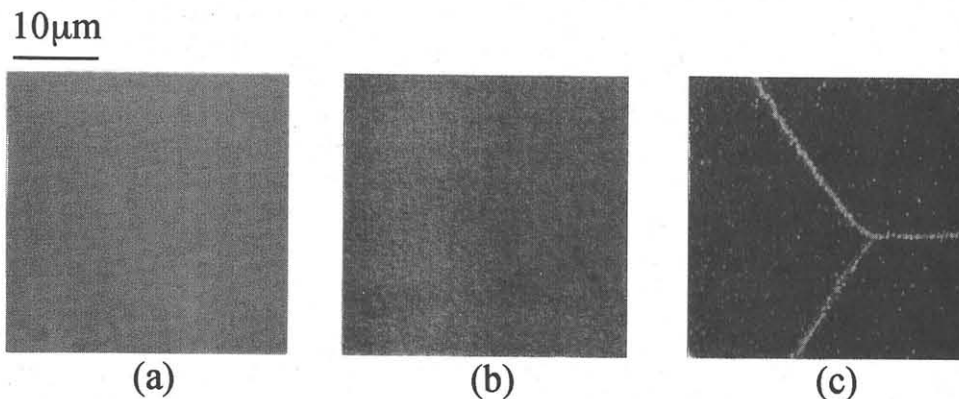


Fig. 4. Surface morphologies of GaN epitaxial layers grown on (a): (0001) Al_2O_3 , (b): (001) MgO, and (c): (001) GaAs substrates.

4. CONCLUSIONS

The GaN growth by RF-radical MBE was investigated. The influence of growth rate and GaN annealed buffer layer on two dimensional growth of hexagonal GaN layers were clarified. That is, the lower growth rate than $0.1 \mu\text{m}/\text{h}$ was required for 2D growth of GaN without buffer layer. The introduction of an annealed GaN buffer layer with 450 W higher RF input power drastically improved the 2D growth behavior of GaN layer on (0001) Al_2O_3 , (001) MgO and (001) GaAs substrates. The dependence of crystal quality of GaN on the difference of substrate materials were described.

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