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Either step-flow or layer-by-layer growth for AlN on SiC (0001) substrates

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ABSTRACT

AlN was grown on 4H- or 6H-SiC (0001) on-axis substrates by plasma-assisted molecular beam epitaxy. By utilizing optimized SiC surface pretreatment, RHEED oscillations just after the growth of AlN were obtained with high reproducibility. This study focused on the growth kinetics of AlN and the correlation between kinetics and the crystalline quality of the grown layers. It was found that the growth mode changed from layer-by-layer to step-flow for high growth temperatures, while for lower temperatures the layer-by-layer growth mode persisted. The mechanism responsible for the change in growth mode is discussed. Symmetrical (0002) and asymmetrical (01-14) x-ray rocking curve measurements were carried out to evaluate the crystalline quality. For the (0002) peak, both high-temperature and low-temperature grown layers showed almost the same FWHM values. On the other hand, for the (01-14) peak, the FWHM of low-temperature grown AlN was much smaller (180 arcsec) than that of the high-temperature grown AlN (450 arcsec).

INTRODUCTION

Growth of high-quality AlN on SiC is a key issue to realize high-performance electronic devices such as GaN-based high-power high-frequency heterojunction field-effect transistors (HFETs) or AlN/SiC metal-insulator-semiconductor FETs (MISFETs). We have investigated precise control of the SiC (0001) surface and optimization of AlN growth conditions in molecular-beam epitaxy (MBE) [1]. Recently, we have realized RHEED intensity oscillations just after the growth of AlN on SiC with high reproducibility [2]. In this study, we focused on the growth kinetics of AlN and the correlation between kinetics and the crystalline quality of the grown layers.

EXPERIMENTAL

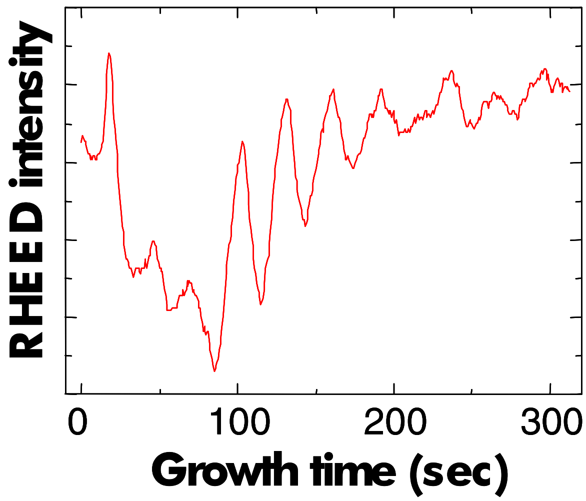
The substrates used in this study were commercially available on-axis 4H- and 6H-SiC(0001)$_{Si}$ wafers (unintentional off angle ~ 0.2°). The substrates were first degreased using conventional organic solvents and dipped in HCl, HCl+HNO_3 (3:1) and HF solutions, and then loaded into a SiC chemical vapor deposition (CVD) system for HCl-gas etching. The HCl-gas etching was carried out at 1300°C for 10 min under a gas flow of HCl (3 sccm) diluted with H_2 carrier gas (1 slm). The details of HCl-gas etching have been reported by Nakamura *et al.*[3] The gas etching procedure successfully removes the polishing scratches typically present on the surfaces of as-received wafers, resulting in an atomically flat terrace structure with a 4 (or 6) monolayer (ML)-height step. Since the atomic stacking of AlN (2H, wurtzite) is different from that of SiC (4H or 6H), formation of steps of unit cell height in the SiC substrate is very important to avoid stacking mismatch boundaries (SMBs) in grown AlN layers. After the HCl-gas etching, the substrates were dipped in an HF solution to remove the native oxide before loading into the MBE system.

The MBE system is equipped with effusion cells for Ga and Al evaporation and an EPI Unibulb radio-frequency (rf) plasma cell for producing active nitrogen. In the MBE chamber, Ga metal was deposited on the substrate at 500-800°C, and then the substrate was heated up to 1000°C to desorb the deposited Ga metal. This procedure was intended to remove residual oxygen atoms from the SiC surface in the form of volatile Ga_xO_y compounds. After the *in-situ* cleaning process, the reflection high-energy electron diffraction (RHEED) patterns were ($\sqrt{3}\times\sqrt{3}$)R30° attributed to 1/3ML-Si adorbed Si-terminated SiC surface [4,5].

AlN growth was carried out at 600-1000°C. A N_2 flow rate of 0.5 sccm and an rf power of 200 W were employed. The Al flux was adjusted very carefully to obtain nearly stoichiometric conditions. The growth rate under this condition was 100 nm/h.

RESULTS AND DISCUSSION

By employing the procedure described above, RHEED intensity oscillations just after the growth were observed with high reproducibility [2]. Figure 1 shows evolution of the RHEED intensity for AlN grown at 1000°C. RHEED intensity oscillations indicating layer-by-layer growth of AlN lasted 10 cycles. Further adjustment of the Al/N ratio did not increase the number of cycles, suggesting the decay of the RHEED oscillations resulted from essential growth physics. As shown in Fig. 2, a 1ML-height step-and-terrace structure was observed for a 60-nm-thick epilayer by atomic force microscopy (AFM). No two-dimensional (2D) nuclei were observed on the grown surface. Therefore, the disappearance of the RHEED oscillations originated from a change in the growth mode, i.e., from layer-by-layer to step-flow growth.



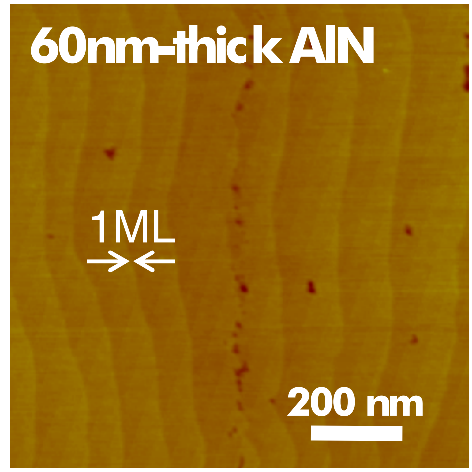


Figure 1. Evolution of the RHEED intensity just after the growth of AlN on a surface-controlled SiC substrate at 1000°C.

Figure 2. AFM image for 60-nm-thick AlN grown at 1000°C. The step height is 1ML.

The change in the growth mode can be explained as follows. The off-cut angle of the SiC substrate is around 0.2°. The terrace width of the initial SiC surface (with 4ML-height step) is 300 nm. A large terrace width results in sufficient supersaturation of adatoms for 2D nucleation on the terrace and thus layer-by-layer growth occurs as illustrated in Fig. 3 (a). As growth proceeds, the 2D nuclei become larger and some of them reach the 4ML-height step-edge and coalesce with it. At that time, the lowest step gains a high effective advance rate as shown in Fig. 3 (b). This is the driving force responsible for dissolving the 4ML-height steps into 1ML-height steps. Finally, the height of all steps becomes 1ML and the terrace width become 4 times smaller than that of the initial surface, 50 nm. For such narrow terrace width, all adatoms are incorporated at the step edge without forming 2D nuclei, i.e., step-flow growth

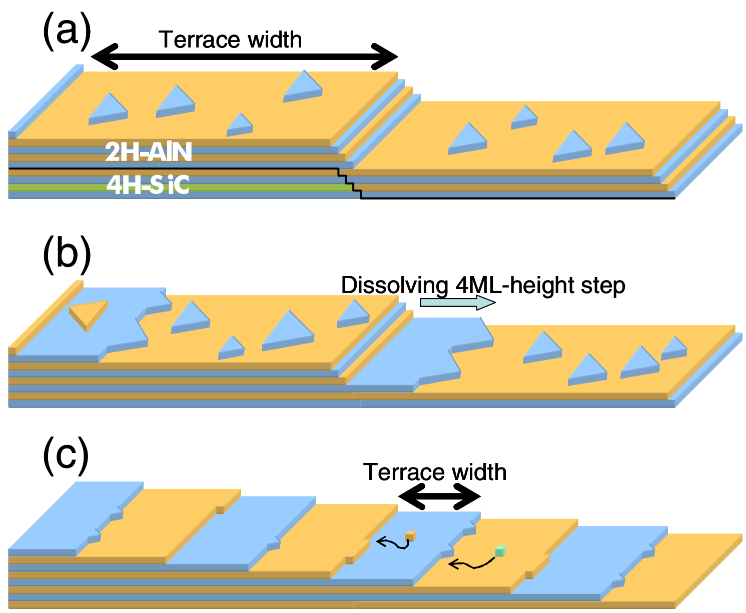


Figure 3. Schematic illustration for growth evolution of AlN. Growth mode changes from layer-by-layer (2D nucleation) to step flow mode.

occurs as shown in Fig. 3 (c). This explanation was confirmed by AFM measurement of epilayers with various thicknesses.

When AlN was grown at a low temperature (600°C), RHEED intensity oscillations lasted over 60 cycles as shown in Fig. 4. Low-temperature growth leads to higher supersaturation and/or shorter migration lengths, which results in such a growth mode. It should be noted that further lowering of the growth temperature resulted in formation of Al dendrites.

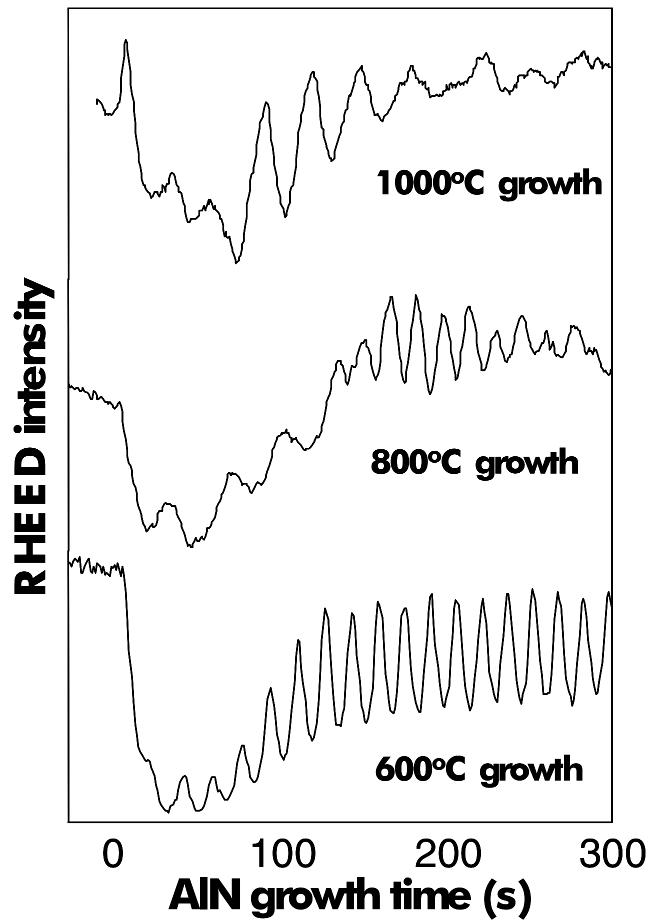


Figure 4. Evolution of RHEED intensity just after the growth of AlN with various growth temperatures.

Symmetrical (0002) and asymmetrical (01-14) X-ray rocking curve (XRC) measurements were carried out for evaluation of the crystalline quality. The results are shown in Fig. 5. For comparison, AlN grown on conventional SiC surface are shown. In this case, AlN grew 3-dimentionally (3D) at the initial stage of growth. As clearly seen, initial 2D growth results in much superior crystalline quality of AlN. Details were discussed in Ref. 2. Now, we focus on 2D-grown samples. For the (0002) peak, both the high-temperature and low-temperature grown layers showed almost the same FWHM values (~70 arcsec for 100 nm-thick AlN). On the other hand, for the (01-14) peak, the FWHM of low-temperature grown AlN was much smaller (180 arcsec) than that of high-temperature grown AlN (450 arcsec). Symmetrical XRC measurements reflect only the density of screw-type dislocations, while asymmetrical XRC measurements are sensitive to both screw- and edge-type dislocations. Thus these results suggest that low-temperature growth resulted in a reduced edge dislocation density. One possible explanation is that layer-by-layer growth leads to a reduced density of edge-type dislocations. Another possible explanation is that the thermal energy in low temperature growth is insufficient to overcome the activation barrier for edge dislocation formation.

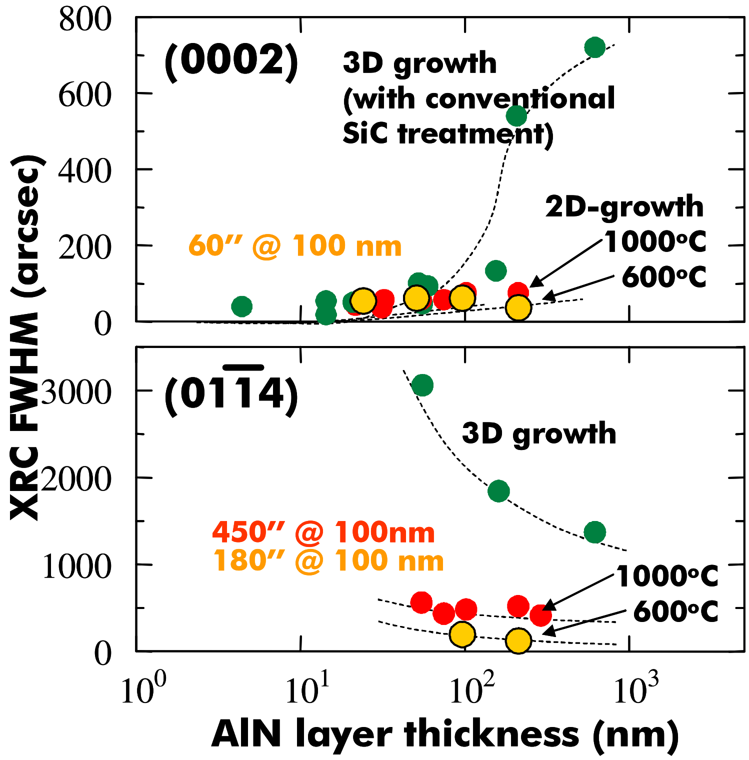


Figure 5. FWHM of symmetrical (0002) and asymmetrical (01-14) x-ray rocking curves for AlN epilayers of various thicknesses.

CONCLUSION

AlN was grown by rf plasma-assisted MBE. In the case of high-temperature growth (1000°C), RHEED intensity oscillations indicating layer-by-layer growth of AlN lasted for only 10 cycles, followed by a transition to step-flow growth. The growth mode transition originates from the dissolution of surface steps of 4ML or 6ML-height into 1ML-height steps. In the case of low-temperature growth (600°C), RHEED intensity oscillations lasted over 50 cycles, indicating that AlN grew in the layer-by-layer mode throughout the growth. Low-temperature growth leads to higher super saturation and/or shorter migration lengths, which make it possible for the layer-by-layer growth mode to be maintained. Symmetrical (0002) and asymmetrical (01-14) X-ray rocking curve measurements were carried out. For the (0002) reflection, both high-temperature and low-temperature grown layers showed almost the same FWHM values (~70 arcsec for 100 nm-thick AlN). On the other hand, for the (01-14) reflection, the FWHM of low-temperature grown AlN was much smaller (180 arcsec) than that of high-temperature grown AlN (450 arcsec). Although further investigation is required, one possible explanation is that layer-by-layer growth resulted in a lower density of edge-type dislocations or reduced twist disorder.

ACKNOWLEDGMENTS

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