Effects of 6*H*-SiC surface reconstruction on lattice relaxation of AlN buffer layers in molecular-beam epitaxial growth of GaN

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Growth of GaN on on-axis 6H-SiC $(0001)_{Si}$ substrates with an AlN buffer layer was performed by molecular-beam epitaxy. The effects of SiC surface reconstruction on the lattice relaxation of AlN buffer layers and the crystalline quality of GaN layers were studied. High-temperature HCl-gas etching followed by HF chemical treatment resulted in an atomically flat SiC surface with a 1×1 structure. The AlN layer grown on the surface showed slow lattice relaxation. GaN grown on the AlN buffer layer exhibited the narrowest (0002) x-ray rocking curve of 70 arcsec and 10^7 cm^{-2} screw-type dislocation density, which was two orders of magnitude smaller than that of GaN grown on as-received substrates. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1533855]

AlGaN/GaN heterojunction field-effect transistors (HFETs) on SiC substrates are most promising for use as high-frequency high-power devices. Molecular-beam epitaxy (MBE) was expected to be a suitable technique for the growth of HFET structures because of its atomic-scale control and the high purity of the layer grown. Although high-performance HFETs grown by MBE have been demonstrated,[1–3] growth of high-quality GaN on SiC is still one of the key issues for improving device performance.

We have earlier reported MBE growth of AlN on 6H-SiC substrates pretreated by high-temperature HCl-gas etching, which resulted in an atomically flat SiC surface with $(\sqrt{3}\times\sqrt{3})R30°$ surface reconstruction. The crystalline quality and surface roughness of AlN grown layers were greatly improved compared to those of AlN grown on an as-received SiC substrate, which suggests that HCl-gas etching is an effective pretreatment for growth of high-quality AlN.[4,5]

In this study, the AlN layer was used as a buffer layer for GaN growth. The growth process of the AlN buffer layer was studied by *in situ* reflection high-energy electron diffraction (RHEED) observation and the quality of GaN layer grown was characterized by atomic force microscope (AFM) and x-ray diffraction (XRD). It was revealed that the surface reconstruction of 6H-SiC has strong effects on the lattice relaxation process of AlN buffer layers and the crystalline quality of GaN.

AlN and GaN layers were grown by plasma-assisted MBE using elemental Ga and Al, and active nitrogen (N*) generated by an EPI Unibulb radio-frequency plasma cell. Commercially available on-axis 6H-SiC $(0001)_{Si}$ substrates were used. After surface pretreatment (described below), the substrate was loaded into the MBE system. Thermal cleaning at 1000 °C for 30 min in an ultrahigh vacuum was followed by growth of 60-nm-thick AlN at 1000 °C under nearly stoichiometric conditions, which resulted in a smooth growth surface. And then, 1-μm-thick GaN was grown at 850 °C under slightly Ga-rich conditions. The growth rates of AlN and GaN were 0.36 and 0.5 μm/h, respectively.

In this study, three different pretreatments for 6H-SiC substrates were investigated. They were combinations of two methods, a wet chemical process and high-temperature HCl-gas etching. First, all substrates were cleaned by a wet chemical process: dipping into aqua regia, HCl solution and HF solution for removal of contamination and natural oxide. These substrates are referred to as the as-received substrates. Many scratches from polishing were observed by AFM on the as-received substrate. Second, some of the substrates were etched by HCl/H_2 gas at 1300 °C to remove the damaged layer caused by polishing, which are referred to as the HCl-treated substrates. The surface exhibited a clear step-and-terrace structure with no scratches. Most of the steps were six monolayers (MLs) in height.[6] The terrace widths were 200–600 nm, corresponding to misorientation of 0.1–0.4°. Finally, some of the HCl-treated substrates were successively cleaned by the same wet chemical process described above. These substrates are referred to as the HF/HCl-treated substrates. The step-and-terrace structure was also observed on the substrate, indicating that the wet chemical process does not affect the surface flatness.

Figure 1 shows RHEED patterns of three 6H-SiC substrates after thermal cleaning in the MBE system. A diffused streaky pattern was observed on the as-received substrate, while sharp intense streaky patterns with clear Kikuchi lines were observed on the HCl-treated and HF/HCl-treated substrates. The as-received substrate exhibited a 1×1 structure. On the HCl-treated substrate, $(\sqrt{3}\times\sqrt{3})R30°$ surface reconstruction, which was thought to be a 1/3 ML Si adatom, was evident,[4,7] whereas a 1×1 structure was observed on the HF/HCl-treated substrate. The Si adatoms may have been removed by the wet chemical process after HCl-gas etching.

AlN buffer layers were grown on these surfaces. Just after the growth of AlN, the RHEED pattern became faint. Then, a streaky pattern appeared gradually. The evolution of *a*-axis lattice constant calculated from the spacing of the streaks is shown in Fig. 2. The lattice constant of bulk (free-standing) AlN is shown by the dashed line. During the initial stage of growth, the lattice constant could not be calculated

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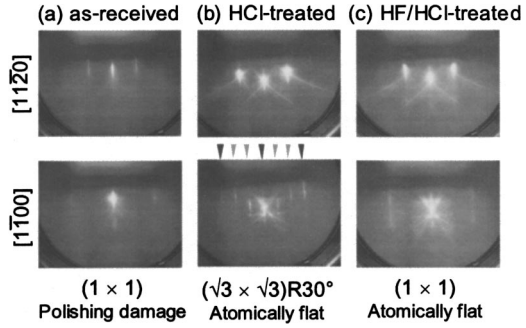

FIG. 1. RHEED patterns of 6H-SiC substrates after thermal cleaning. (a) As-received substrate, (b) HCl-treated substrate and (c) HF/HCl-treated substrate. Intense streaky patterns with Kikuchi lines are observed in (b) and (c), indicating an atomically flat surface. No reconstruction is observed in (a) and (c), whereas $(\sqrt{3}\times\sqrt{3})R30°$ reconstruction is observed in (b).

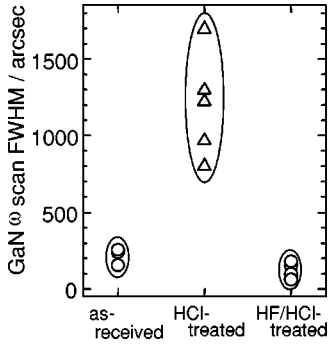

FIG. 3. Full widths at half maximum of x-ray rocking curves for (0002) diffraction from GaN layers grown on 6H-SiC substrates with three different pretreatments: (a) the as-received substrate, (b) the HCl-treated substrate and (c) the HF/HCl-treated substrate. The GaN layer grown on the HCl-treated substrate exhibits considerably wider peaks compared to the others.

because of the faint RHEED pattern. In all cases, lattice relaxation due to the lattice mismatch between SiC and AlN was observed, but the relaxation processes were different. On the as-received substrate, the lattice constant increases slowly. Even after 600 s (60 nm) growth, the lattice constant still remained in the middle of bulk AlN and SiC, indicating that the AlN layer has compressive strain. On the other hand, the lattice constant is close to that of bulk AlN within 200 s (20 nm) on the HCl-treated substrate, indicating full relaxation of the AlN layer. The evolution of the lattice constant on the HF/HCl-treated substrate was similar to that observed on the as-received substrates, but the AlN layer has more compressive strain. These results clearly indicate that the strain relaxation process of AlN was strongly affected by 6H-SiC substrate pretreatment. It is thought that the SiC initial surface with $(\sqrt{3}\times\sqrt{3})R30°$ resulted in fast relaxation of the AlN layer.

A main GaN layer was grown after grown of the AlN buffer layer. During and after the growth of GaN, RHEED exhibited a streaky pattern for all samples. The full widths at half maximum (FWHMs) of the GaN (0002) x-ray rocking curve (XRC) measurements are plotted in Fig. 3. To check the reproducibility, at least three samples were grown on individual substrates. As is clearly seen, the FWHMs on the HCl-treated substrate range from 700 to 1700 arcsec, significantly larger than those on the other two substrates, indicating poor crystalline quality of GaN. The GaN grown on the as-received substrates exhibits relatively good values, 150–250 arcsec. Surprisingly, on the HF/HCl-treated substrates, the best value obtained was 70 arcsec, which is a considerably small value for MBE-grown GaN on SiC.[8,9]

Figure 4 shows AFM images of GaN layers using two different scan sizes, 20 and 1 μm. On the as-received substrate, a lot of hillocks are observed in the 20 μm scan image. As shown in the 1 μm scan image of Fig. 4(a), these hillocks originate from spiral growth at screw-type (pure screw or mixed) dislocations.[10] The density of the hillocks was $\sim 10^9$ cm^{-2}. On the HCl-treated substrate, irregular steps and small holes are observed [Fig. 4(b)], showing the poor crystalline quality of GaN. On the HF/HCl-treated substrate, hillocks were also observed (as bright spots in the 20 μm scan image), but the density was $\sim 10^7$ cm^{-2}, two orders of magnitude smaller than that on the as-received substrate. Between hillocks, a clear step-and-terrace structure is observed in the 1 μm scan image in Fig. 4(c). These results suggest that HF/HCl treatment resulted in a remarkable reduction of screw-type dislocations in GaN grown layers. However, the presence of dark spots in the AFM image may indicate the presence of other dislocations. The reduced density of dislocations needs to be confirmed by transmission electron microscopy.

Although both HCl-treated and HF/HCl-treated SiC substrates have a very flat surface, the crystalline quality of GaN

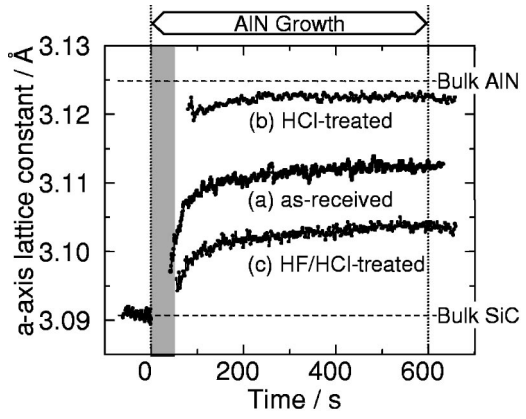

FIG. 2. Evolution of the *a*-axis lattice constant during AlN buffer layer growth on three different substrates: (a) the as-received substrate, (b) the HCl-treated substrate and (c) the HF/HCl-treated substrate. The change in *a*-axis lattice constant indicates lattice relaxation of the AlN layer.

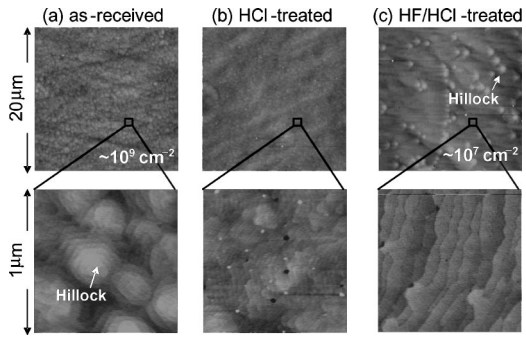

FIG. 4. AFM images of GaN layers grown on 6H-SiC substrates with three different pretreatments: (a) the as-received substrate, (b) the HCl-treated substrate and (c) the HCl/HF-treated substrate. Spiral growth hillocks are observed in (a) and (c), whereas no clear structure is observed in (b). The density of the hillocks in (c) is two orders of magnitude smaller than that in (a).

grown on these substrates was very different as discussed above. These two SiC substrates have different surface reconstruction, which resulted in the different relaxation process for the AlN buffer layer. Detailed studies on the crystalline quality of the AlN buffer layer will be key to understanding why the quality of GaN is so different. Another possible explanation is strain. There is 2.4% lattice mismatch between GaN and AlN. Strain relaxation occurs during GaN growth on AlN. If AlN has compressive strain, the mismatch becomes larger. It may be possible that the stress at the GaN/AlN interface results in bending of the dislocation or annihilation of two dislocations with opposite Burgers vectors.

In summary, we proposed high-temperature HCl-gas etching followed by chemical treatment as a SiC substrate pretreatment, which resulted in 1×1 reconstruction with an atomically flat surface. On the pretreated SiC substrate, the AlN layer showed slow lattice relaxation. AlN had compressive strain even after 60 nm thick growth. GaN layers grown on the AlN buffer layer exhibited the narrowest XRC (0002) diffraction of 70 arcsec and 10^7 cm^{-2} screw-type dislocation density, which is two orders of magnitude smaller than that of GaN grown on as-received substrates.

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