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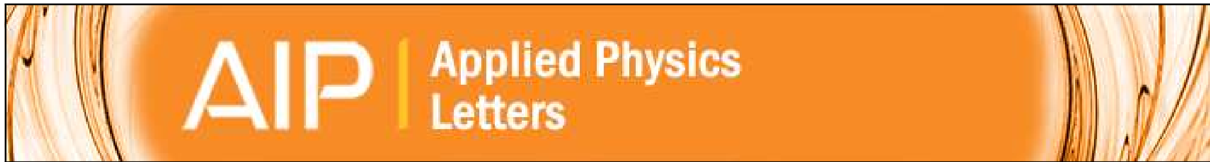

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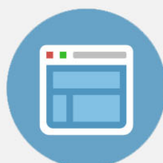

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Initial stages of molecular-beam epitaxy growth of GaN on 6H-SiC(0001)

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We studied the atomic H etching of 6H-SiC substrates and the initial stages of GaN/6H-SiC molecular-beam epitaxy growth. Atomic H etched 6H-SiC$(0001)_{Si}$ and $(000\bar{1})_C$ surfaces show a $(\sqrt{3}\times\sqrt{3})-R30°$ and a (1×1) reconstruction respectively, with 0.7 ± 0.2 monolayers of remnant O on both surfaces. GaN/6H-SiC$(0001)_{Si}$ growth is initiated by the formation of islands that develop into flat-top terraces through coalescence. Growth steps of one or integer numbers of the GaN atomic bilayer height are observed. GaN grown on 6H-SiC$(000\bar{1})_C$ is rougher with islands of irregular shape. X-ray photoemission spectroscopy studies show that Si $2p$ and C $1s$ photoelectron inelastic mean free paths in GaN are 22 ± 1 and 20 ± 1 Å, respectively. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1350430]

The wide-band-gap semiconductor GaN can now be successfully grown and used for fabricating blue light emitting and laser diodes.[1,2] Although the most commonly used substrate is sapphire, 6H-SiC is considered a promising alternative. In addition to being more closely lattice matched to GaN (lattice mismatch ~3.4%), 6H-SiC is a good electrical and thermal conductor, factors which are highly advantageous in both optoelectronic and high power electronic applications. However, despite the fact that 6H-SiC is better lattice matched to GaN, the quality of GaN grown on 6H-SiC does not yet exceed that of GaN grown on sapphire. This is partly due to the poor understanding of the crucial initial stages of the GaN/6H-SiC growth. In this work, the atomic H etching of the 6H-SiC substrate and the initial stage of GaN/6H-SiC molecular-beam epitaxy (MBE) growth are studied using reflection high-energy electron diffraction (RHEED), x-ray photoemission spectroscopy (XPS), and atomic force microscopy (AFM). The polarities of the GaN films are also studied using chemical etching.

Our home-built MBE system is equipped with an Oxford Applied Research CARS-25 rf plasma gas source and an *in situ* XPS (Mg anode).[3] AFM was performed in tapping mode in air ambient using a Digital Instruments Multimode™ scanning probe microscope. The substrates are Epitronics *n*-type on-axis 6H-SiC$(0001)_{Si}$ and $(000\bar{1})_C$. Following solvent degreasing, the substrates were etched first by H_2SO_4:HNO_3=1:1 for 5 min then by HF:H_2O=1:4 for 10 min before being In mounted onto a Mo block and loaded into the MBE chamber. Subsequently, the substrates were etched at 650 °C using atomic H with a rf power of 400 W and a chamber H_2 pressure of 5×10^{-5} mbar for 60 min.

After atomic H etching, for 6H-SiC$(000\bar{1})_C$, a (1×1) RHEED pattern was observed. For 6H-SiC$(0001)_{Si}$, the pattern was a sharp streaked $(1\times)$ in $[11\bar{2}0]$ direction and a blurred $(3\times)$ in $[1\bar{1}00]$ direction indicative of a $(\sqrt{3}\times\sqrt{3})-R30°$ reconstruction. XPS O $1s$ peak studies showed that atomic H etching at even higher temperature (830 °C) cannot remove O completely. The O coverage on both faces was 0.7 ± 0.2 ML as measured by quantitative XPS. This is in contrast with the report of Lin *et al.*[4] where complete O removal from 6H-SiC surface by atomic H was claimed based on XPS analysis of Si $2p$ peak shape changes. We believe that Si $2p$ peak shape is not very sensitive to the surface O in this case. Nevertheless, the slightly O covered 6H-SiC$(0001)_{Si}$ $(\sqrt{3}\times\sqrt{3})-R30°$ surface has previously been associated with a well-ordered hexagonal structure,[5] and high quality GaN films have been grown on similarly prepared surfaces.[6]

GaN films were grown at 650 °C by applying Ga and N fluxes simultaneously. A slightly Ga rich condition was used, corresponding to a Ga cell temperature of 1030 °C and a chamber N_2 pressure of 5×10^{-5} mbar with a rf power of 220 W. The growth rate was 0.17 ± 0.01 Å/s (~5 times lower than that for normal GaN growth) as calibrated by measuring the thickness of thick films [grown on both $(0001)_{Si}$ and $(000\bar{1})_C$] using both an optical interference method and transmission electron microscopy.

Upon initiation of the growth for GaN/6H-SiC$(0001)_{Si}$, the $(1\times)$ RHEED pattern in $[11\bar{2}0]$ direction remained streaky for a few seconds, then became increasingly spotty indicative of island formation. The spottiest pattern was observed when ~16 Å GaN had been deposited. A streaky RHEED pattern gradually returned as the film grew thicker than ~40 Å suggesting the coalescence of the islands.

The XPS Si $2p$ peak shape change due to the possible nitridation of SiC was not observed for this slightly O covered substrate. XPS intensities (peak area) were analyzed to investigate the growth mode following an established method.[7] In Fig. 1, normalized intensities of substrate Si $2p$ and C $1s$ are plotted as a function of GaN overlayer thickness. After a dip at 0.6 Å, the intensities decrease almost linearly until ~25 Å where the decrease becomes exponential. This can be understood as follows. At 0.6 Å, the film was two dimensional (2D), therefore Si $2p$ and C $1s$ intensities dropped exponentially. Further growth led to formation of GaN islands that consumed atoms from the previously grown film as well as the impinging Ga and N atoms. As a result, a fraction of substrate area became nude giving rise to

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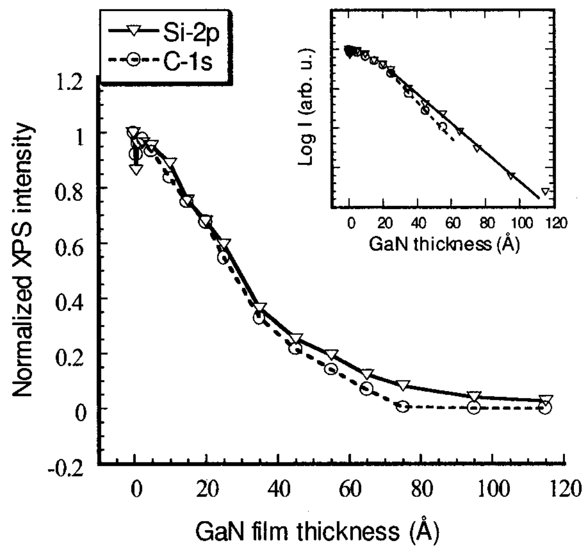

FIG. 1. Normalized substrate Si $2p$ and C $1s$ XPS intensities as a function of GaN film thickness. The inset is the same data plotted on logarithmic scale. The slopes of the straight fitting lines give the inelastic mean free path of Si $2p$ and C $1s$ in GaN.

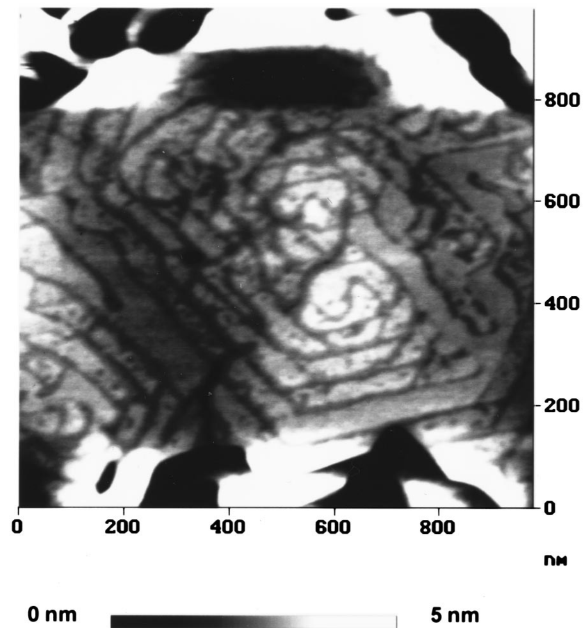

FIG. 3. AFM image of 1100 Å GaN grown on 6H-SiC$(0001)_{Si}$. A large hexagon terrace with growth steps.

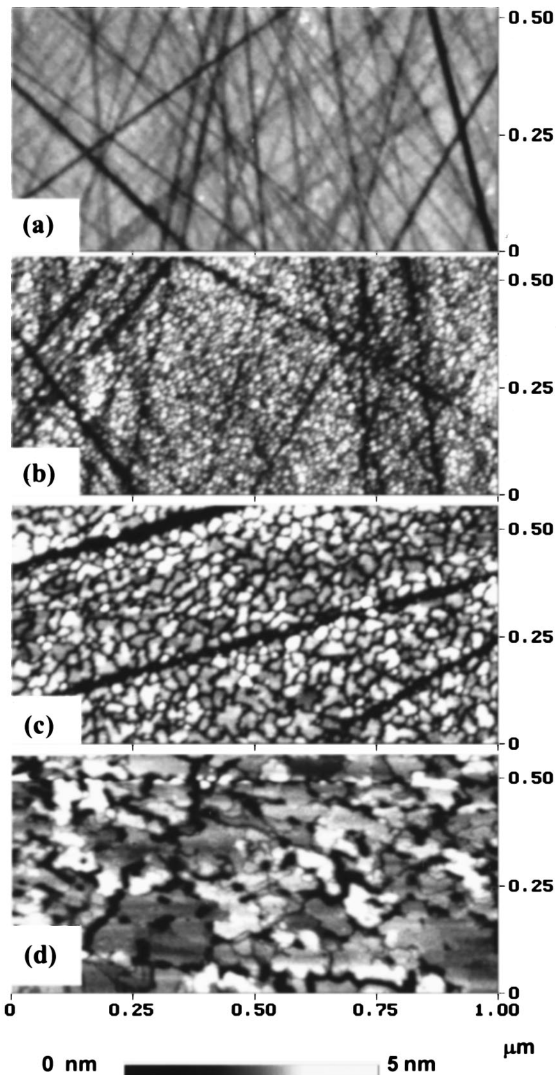

FIG. 2. AFM images of GaN grown on 6H-SiC$(0001)_{Si}$. (a) The substrate (b) 12 Å, (c) 40 Å, and (d) 110 Å.

an increase in substrate photoemission intensity. As those islands grew thicker and wider, the substrate peak intensities decreased but slower than an exponential decay expected for 2D growth. When these islands coalesced after ~25 Å, the growth hitherto was 2D-like, and Si $2p$ and C $1s$ intensities decreased exponentially. This interpretation is consistent with the RHEED observation. From the fit of the data after 25 Å (the inset in Fig. 1), we obtained Si $2p$ and C $1s$ photoelectron (kinetic energy 969.6 and 1152.6 eV) inelastic mean free path in GaN to be 22 ± 1 and 20 ± 1 Å, respectively.

For morphology studies, two samples were grown on 6H-SiC$(0001)_{Si}$, and $(000\bar{1})_C$ using otherwise identical growth conditions. Each growth was interrupted at 12, 40, and 110 Å for AFM measurements. After each AFM measurement, the sample was reloaded and cleaned using atomic H at 650 °C for 10 min. After this cleaning, a (2×2) GaN RHEED pattern was observed; and the surface contamination was below the XPS detectable level. The growth was resumed without introducing too many defects as demonstrated by transmission electron microscopy.

The morphology of the sample grown on 6H-SiC$(0001)_{Si}$ is shown in Fig. 2. Figure 2(a) is the morphology of the as-received substrate. Islands of typical size ~12 nm were observed at 12 Å [Fig. 2(b)]. At 40 Å, these islands grew to a typical size of ~30 nm [Fig. 2(c)], and the shape of the islands became polygon-like with flat tops. These flat-top polygon islands are not usually observed in cubic semiconductor on cubic semiconductor (001) MBE growth where the islands are rounded or faceted.[8–10] This may result from the surface energy anisotropy where the wurtzite GaN surface grown on 6H-SiC$(0001)_{Si}$ has minimum surface energy and the islands prefer a flat-top shape to minimize the total surface energy according to the Wulff construction which can be applied to understand non-equilibrium MBE growth.[11] At this stage, each island makes contact with its neighboring islands giving evidence to the

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coalescence. At 110 Å [Fig. 2(d)], the surface grew flatter with larger coalesced islands. After 1100 Å GaN had been grown (Fig. 3), large hexagonal terraces with growth steps of ~2.5 Å or multiples of 2.5 Å in height were observed consistent with the height of one or an integer number of GaN bilayers. In contrast, GaN grown on $(000\bar{1})_C$ is rougher and consists of islands with irregular shape. The growth steps were not observed.

The polarities of the two samples were determined by chemical etching using a 20% KOH aqueous solution for 5 min at room temperature.[12] The etching resulted in little change on the morphology of the film grown on $(0001)_{Si}$, whereas considerable morphology change was observed for the film grown on $(000\bar{1})_C$. According to Ref. 12, the film grown on $(0001)_{Si}$ and $(000\bar{1})_C$ are assigned to Ga and N polarity, respectively.

In conclusion, 6H-SiC prepared by *in situ* atomic H etching at 650 °C exhibits a weak $(\sqrt{3}\times\sqrt{3})-R30°$ and a (1×1) RHEED pattern for $(0001)_{Si}$ and $(000\bar{1})_C$ face, respectively, with a remnant O coverage of $\sim0.7\pm0.2$ monolayers for both faces. XPS studies show that the inelastic mean free path for Si 2p and C 1s photoemissions in GaN are 22 ± 1 and 20 ± 1 Å, respectively. The GaN/6H-SiC$(0001)_{Si}$ MBE growth is initiated by the formation of islands following 0.6 Å of two-dimensional growth. Further growth results in coalescence of the islands at ~25 Å. The flat-top shaped islands and flat terraces with growth steps of one or integer number of GaN atomic bilayer height are observed by AFM. Whereas GaN grown on 6H-SiC$(000\text{-}1)_C$ is rougher. KOH etching indicates that the GaN grown on 6H-SiC$(0001)_{Si}$ is of Ga polarity, and the GaN grown on 6H-SiC$(000\bar{1})_C$ is of N polarity.

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