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## Structural anisotropy in GaN films grown on vicinal 4H-SiC surfaces by metallorganic molecular-beam epitaxy

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GaN grown directly on 4H-SiC substrates by metallorganic molecular-beam epitaxy is investigated in terms of nucleation, coalescence, and growth front evolution. The effects of SiC surface configurations such as step and terrace structures on GaN film growth physics are examined in detail. Comparative studies using on-axis and vicinal SiC surfaces indicate distinguishable differences in structural and morphological characteristics. An anisotropic x-ray characteristic is observed for the GaN film deposited on the vicinal stepped SiC surfaces. This is due to preferential nucleation and coalescence of GaN islands along step edges, which are induced by the confinement of adatoms on the narrow terraces on the vicinal SiC surface. © 2003 American Institute of Physics. [DOI: 10.1063/1.1605791]

One difficulty in manufacturing GaN-based devices is the lack of native GaN substrate material. To overcome this, group III nitride semiconductors have been grown on sapphire substrates using a low temperature buffer layer technique.<sup>1</sup> The film quality has been improved enough to achieve commercial light emitting diodes.<sup>2</sup> Laser diodes have also been commercialized by the use of epitaxial lateral overgrowth, which enables a reduction of the density of threading dislocations.<sup>3</sup> However, a lot of attention has been paid to SiC, another possible substrate material candidate for growth of high-quality GaN. The properties of SiC are superior to those of sapphire with respect to, for example, lattice mismatch and thermal conductivity. Thus, epitaxial growth by using SiC as substrate material should potentially produce higher quality nitride films than growth on sapphire substrates. However, the resultant films are of a quality comparable to those grown on sapphire mainly because of undefined SiC substrate surfaces consisting of scratches and nanometer scale surface roughness.

Recently, some groups<sup>4-7</sup> have studied heteroepitaxial growth mechanisms with the use of atomically smooth SiC surfaces, which were obtained either by H<sub>2</sub><sup>8,9</sup> or HCl/H<sub>2</sub><sup>10</sup> gas etching at high temperatures. We have previously reported the defect formation mechanism in AlN thin films on stepped 6H-SiC surfaces during metallorganic chemical vapor deposition growth.<sup>4</sup> AlN nuclei formed on an atomically smooth SiC surface coalesce without any defect formation due to the coherent relationship between the nuclei. Incoherent boundaries such as double positioning boundaries<sup>11</sup> are, on the contrary, introduced at the coalescence stage of AlN nuclei which are formed on different terraces. The incoherent nature of nuclei induces kinetic/energetic barriers for the diffusion of adatoms and thus causes the delay of coalescence of nuclei. Cheung *et al.* showed the evolution of GaN nuclei on stepped SiC surfaces by scanning tunneling microscopy during molecular-beam epitaxy (MBE) growth.<sup>7</sup>

In this work, we have studied GaN nucleation and

growth on vicinal SiC surfaces. We found that the growth progresses with a distinct terrace width, which is uniquely determined by the initial cut angle of the wafer.<sup>12</sup>

GaN growth was performed on commercially available 4H-SiC substrates by metallorganic MBE using methylhydrazine as a nitrogen source and Ga metal. The substrates used in this study were both on-axis and vicinal ( $\sim 8^\circ$  off toward  $[1\bar{1}20]$ ) 4H-SiC(0001) (Si face). Prior to GaN growth the substrate was thermally annealed at 1430 °C in the H<sub>2</sub>/HCl gas for 10 min to obtain atomically smooth surfaces. The surface was examined by atomic force microscope (AFM), showing clear straight steps and terrace configurations. Due to energetically driven step-bunching the step height on vicinal 4H-SiC surfaces always appeared to be 1 nm, corresponding to 1-unit-cell,<sup>12</sup> and the terrace width was  $\sim 160$  nm for on-axis and  $\sim 9$  nm for vicinal substrates.

GaN films with a thickness of  $\sim 150$  nm were grown simultaneously on these substrates at 675 °C (pyrometer reading) for 1 h. *In situ* reflection high-energy diffraction showed a streaky (1×1) pattern, indicative of a smooth surface. The resultant films were examined by x-ray rocking curve (XRC) measurement of the GaN (0002) and transmission electron microscopy (TEM) using a Topcon EM-002B.

Figure 1 show the XRC results on both samples. The full width at half maximum (FWHM) value of the XRC ( $\omega$ -scan) measurement of the on-axis sample is  $\sim 365$  arc sec from both x-ray incident directions of  $[1\bar{1}20]$  and  $[1\bar{1}00]$ , which are, respectively, perpendicular and parallel to step edges, indicating GaN film quality used in this study is comparable to of generally obtained GaN films by MBE<sup>7</sup> and structurally isotropic characteristic. In contrast, the vicinal sample show an anisotropic characteristic as can be seen in the rocking curves taken from  $[1\bar{1}20]$  and  $[1\bar{1}00]$  incident x-ray directions (see the schematic figure inset describing x-ray incident directions in relation to surface steps). The FWHM value from  $[1\bar{1}00]$  is 162 arc sec, which is clearly smaller than 250 arc sec from  $[1\bar{1}20]$ . This implies a larger concentration of defective features across the step edges, which will also be further investigated in the following section.

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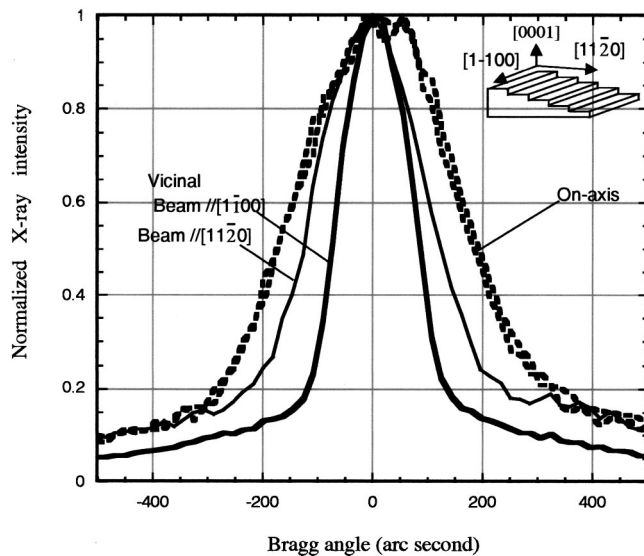


FIG. 1. X-ray rocking curves of GaN(0002) grown on the on-axis and vicinal SiC substrates. An inset figure shows the incident directions of x-ray beam on the vicinal SiC sample. Note the anisotropic feature in the vicinal sample.

Figures 2(a) and 2(b) show cross-sectional bright-field TEM images. Both GaN films reveal defective features but the defect nature is significantly different. The on-axis sample shown in (a) contains threading dislocations as typically seen in GaN thin films. On the contrary, the vicinal sample shown in (b) includes stacking faults parallel to the film surface, as indicated by arrows, in addition to threading dislocations. Importantly some of the threading dislocations are terminated by the stacking faults,<sup>13</sup> probably resulting in smaller x-ray FWHM values than of the on-axis sample.

In order to understand the structural differences we have investigated the nucleation and coalescence stages of GaN on stepped SiC surfaces. Similar substrates and the same growth conditions were used as in the thick GaN growth. The deposition time was the only parameter that was varied, i.e., 40 and 120 s. After 40 s of growth a high density of GaN nuclei was observed, as shown in Figs. 3(a) and 3(b). Notice the differences between the on-axis and the vicinal sample with respect to nucleation sites and its density; random nucleation on the wider terraces is evident on the on-axis sample, whereas one-dimensionally aligned nucleation along step edges is featured in the vicinal sample. The nucleation density on (a) the on-axis and (b) the vicinal substrate is  $\sim 9 \times 10^{10} \text{ cm}^{-2}$  and  $\sim 2.7 \times 10^{11} \text{ cm}^{-2}$ , respectively.

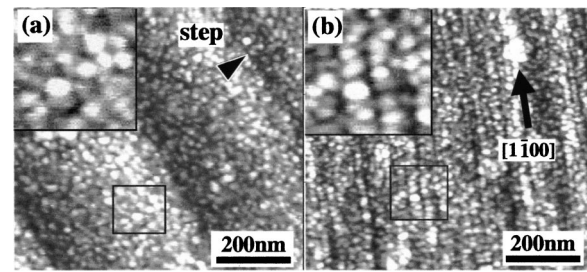


FIG. 3. AFM images of GaN nuclei formed on (a) the on-axis and (b) the vicinal SiC surfaces. Random and one-dimensionally aligned nucleation is evident on (a) the on-axis and (b) the vicinal SiC surface, respectively. Inserted figures show the magnified images of a region indicated by the square.

These differences can be explained by the presence of kinetic asymmetry of adatoms at step sites, the Schwoebel effect,<sup>14</sup> the adatoms on the narrower terrace are effectively confined in one direction towards step site and thus diffuse along step edges. This results in the increase of adatom collision rate, which accordingly induces higher nucleation density. After 120 s of growth GaN nuclei start to coalesce as seen in Figs. 4(a) and 4(b). 3–5 nuclei coalesced to form a circular island with a radius of  $\sim 40 \text{ nm}$  on the on-axis surface. In contrast, the nuclei tend to coalesce along the step edge direction  $[1\bar{1}00]$  and form elongated islands on the vicinal surface. This is a similar behavior that Cheung *et al.* observed for on-axis and off-axis ( $8^\circ$  off) SiC substrates during MBE growth.<sup>7</sup>

Since most defects in GaN film are generated at the coalescence stage of nuclei the structural difference between the on-axis and vicinal samples can be better understood with the help of the AFM images shown in Fig. 4. In both cases the majority of defects is a threading dislocations, which are introduced when two nuclei coalesce.<sup>15–17</sup> If the nuclei are incoherent and, in most cases, probably also twisted/tilted with respect to each other due to a mismatch relaxation process, the edge-type dislocation having a Burgers vector parallel to the basal plane is generated at the boundary to compensate incoherent lattices. The plan-view TEM study on GaN nuclei on SiC surfaces indicates a wandering feature of moiré fringes and the presence of dislocations as a result of incoherent nature of GaN nuclei.<sup>18</sup> The degree of incoherency and twist/tilt of neighboring nuclei is determined by their size and density.<sup>7</sup> The bigger the island size is the more twist/tilt of neighboring nuclei is. This is likely to be an effect of energy minimization since the elastic energy in a

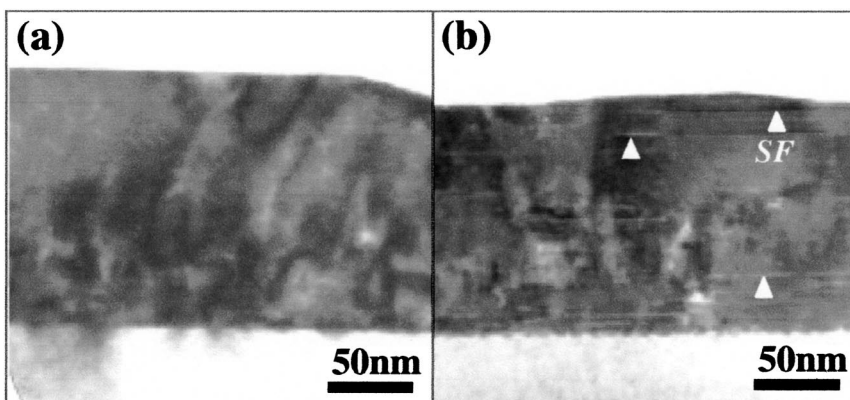


FIG. 2. Bright-field cross-sectional TEM images of GaN films on (a) the on-axis and (b) the vicinal SiC substrates. The arrows in (b) indicate some of the stacking faults.

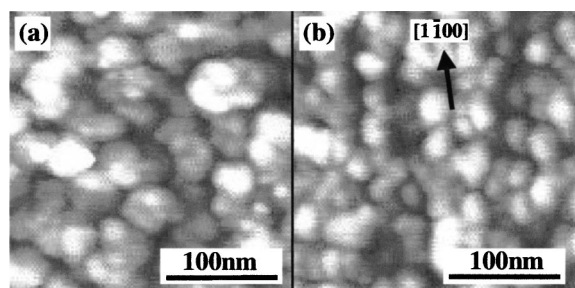


FIG. 4. AFM images of GaN coalesced islands formed on (a) the on-axis and (b) the vicinal SiC surfaces. Circular and elongated islands after coalescence are seen.

nucleus is probably reduced by twisting/tilting motion. This motion is more pronounced among the randomly nucleated islands on the on-axis surface than among the one-dimensionally nucleated islands on the vicinal surface. Earlier coalescence caused by a higher nucleation density giving rise to less twist/tilt means less energetic relaxation in a nucleus, which in turn keeps the coherency of the nuclei.<sup>7</sup> In this way the defect density in  $[1\bar{1}00]$  direction is kept smaller than in  $[11\bar{2}0]$  direction, in which more defects are introduced when islands coalesce over the step edges.<sup>11</sup>

In summary, different stages of GaN growth directly on 4H-SiC substrates by metallorganic molecular-beam epitaxy were investigated. The SiC surfaces used in this study were specially etched at high temperature to achieve atomically smooth terrace and step structures. A crystallographic anisotropy present in the thin GaN film deposited on the vicinal stepped SiC surface was investigated with respect to nucleation and coalescence. AFM observations of the nucleation and coalescence stages clearly showed distinguishable differences in GaN nucleation and coalescence behavior between the on-axis and vicinal surfaces; random and one-dimensionally aligned nucleation was observed on the on-axis and vicinal surfaces, respectively. An early coalescence

due to a higher nucleation density and preferential coalescence along step edges was found to substantially improve the GaN crystal quality of the film grown on the vicinal SiC surface.<sup>17</sup>

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