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Defect structure in selective area growth GaN pyramid on (111)Si substrate

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A GaN pyramid grown selectively on a (111)Si substrate with a patterned dot structure of a SiO_2 mask, by metalorganic vapor phase epitaxy using AlGaN as an intermediate layer, was characterized by transmission electron microscopy. The dot pattern has an array of 5.0-μm-diameter window openings with a 10 μm period. The density of threading dislocations observed in the window region decreased gradually with increasing distance from the interface. This was mainly due to the dislocation reaction and bending of threading dislocations for the first 2 μm region from the interface and for the upper region, respectively. Dominantly observed defects in the lateral-growth part were dislocations parallel to the interface. An amorphous layer was formed at the interface in the window region. Nitride particles were observed at the interface in the mask region. *© 2000 American Institute of Physics.* [S0003-6951(00)01619-3]

Selective area growth (SAG) of GaN is a promising technique for the fabrication of optical and electrical devices with high performance. Reduction of the threading dislocation density has been reported for SAG of GaN on sapphire[1,2] and 6H-SiC[3] substrates. Recently, Si has been considered as an alternative substrate because of its low cost, large size, and the potential for integration of GaN-based devices with Si-based ones. To obtain SAG of GaN, Mao *et al.*[4] and Marchand *et al.*[5] have used predeposited GaN/AlN on (111)Si and predeposited AlN on (111)Si as substrates, respectively. Also, Kobayashi *et al.*[6] have demonstrated SAG of GaN using $AlO_x/(111)$Si substrates. All of them confirmed a reduction of the threading dislocation density, however their growth processes are performed in two steps. On the other hand, Kawaguchi *et al.*[7] have demonstrated, by metalorganic vapor phase epitaxy (MOVPE), SAG of GaN directly on (111)Si using AlGaN as an intermediate layer. In their growth, by introducing AlGaN, the formation of island-like structures of AlGaN occurs on Si. The AlGaN islands play the role of nucleation centers for the growth of GaN. The initial growth of GaN is enhanced at the edge of the window region. By continuing the growth of GaN further, the neighboring islands of GaN begin coalescing with one another, and micron-sized pyramids of GaN are formed. To obtain such a structure, the selection of the composition of AlGaN is very important. When an AlN-rich composition is used, AlGaN islands occur in the mask region as well as in the window region, while growth of GaN does not occur when AlGaN is not used. The AlN mole fraction for successfully obtaining SAG of GaN is estimated to be 0.09 by x-ray diffraction measurements using AlGaN grown on a nominally unpatterned (0001) sapphire substrate. In this letter, we present transmission electron microscopy (TEM) characterization of the GaN pyramid on (111)Si.

The GaN pyramids used in this study were grown by MOVPE, using AlGaN as an intermediate layer, on (111)Si substrates with a patterned dot structure of a silicon dioxide (SiO_2) mask. Patterning of the mask was carried out by conventional photolithography and dot-patterned windows were formed by wet etching. The dot pattern has an array of 5.0-μm-diameter window openings with a 10 μm period. Trimethylgallium, trimethylaluminum, and ammonia (NH_3) were used as source gases of Ga, Al, and N, respectively. The AlGaN intermediate layer was grown on the dot-patterned Si substrate at 1090 °C, and then GaN was grown at 1090 °C. Details of the growth procedure are described in Ref. 7. Cross-sectional TEM samples were prepared by mechanical prethinning followed by final thinning using low-angle, 3–5 kV, Ar-ion milling. The observations were carried out using electron microscopes; Hitachi H-8000 operated at 200 kV and Hitachi H-1250ST operated at 400 or 1000 kV. The composition of AlGaN was determined by energy dispersive x-ray (EDX) spectroscopy. The measurements were performed using a probe size of approximately 20 nm, which is smaller than the size of AlGaN islands (0.1–0.5 μm).

First, the epitaxial relationships between the nitride and Si were determined by selected area electron diffraction. The location of AlGaN was determined by EDX. The result indicates that the nitride has a wurtzite structure and an orientation relationship as follows: $[11\bar{2}0]$GaN//$[11\bar{2}0]$AlGaN//$[1\bar{1}0]$Si and [0001]GaN//[0001]AlGaN//[111]Si. These findings are consistent with previously published results.[8] In the following, the observations were carried out along the $[11\bar{2}0]$ zone axis of the nitride.

Figures 1(a) and 1(b) show typical low-magnification

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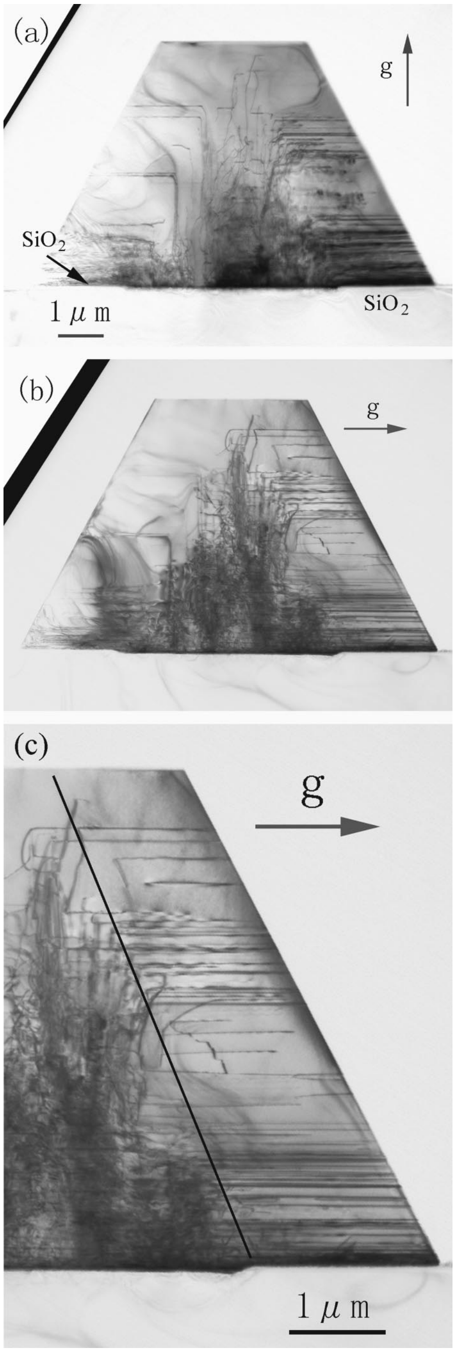

FIG. 1. Cross-sectional TEM images of GaN pyramid structure. (a) $\mathbf{g}=[0002]$, (b) $\mathbf{g}=[1\bar{1}00]$, (c) enlarged image of the right-hand side of the pyramid of (b).

bright-field TEM images of a pyramid structure imaged with $\mathbf{g}=[0002]$ and $\mathbf{g}=[1\bar{1}00]$, respectively. It is clearly seen that threading dislocations are generated at the interface region in the window where GaN was grown on Si using the AlGaN intermediate layer. These dislocations are introduced by coalescence of the nitride islands and by lattice mismatch between the nitride and Si. However, no newly created threading dislocations are observed at the interface between the nitride and the mask, indicating that lateral growth occurred in the mask region. The density of the threading dislocations is very high near the interface region in the window ($\sim 10^{10}\,\mathrm{cm}^{-2}$), but it decreases gradually with increasing distance from the interface. The reduction of the threading dislocation density appears to mainly be due to dislocation reactions[9–11] within the first $\sim 2\ \mu$m of the window region. Above this region where little interaction between dislocations occurs, the reduction of the threading dislocation density is mainly caused by bending of threading dislocations.[1–3] In Fig. 1 one can clearly see that some of the threading dislocations bend, and then propagate parallel to the interface toward the outer regions.

The type of threading dislocation is determined by comparing Figs. 1(a) and 1(b). In general, threading dislocations with Burgers vectors $\mathbf{b}=[0001]$, $\mathbf{b}=1/3[11\bar{2}0]$, and $\mathbf{b}=1/3[11\bar{2}3]$ have been observed in epitaxially grown GaN.[10–15] As can be seen, almost all threading dislocations visible for $\mathbf{g}=[0002]$ are also visible for $\mathbf{g}=[1\bar{1}00]$, indicating that the threading dislocations observed in Fig. 1(a) are of the $\mathbf{b}=1/3[11\bar{2}3]$ type. Other threading dislocations visible only for $\mathbf{g}=[1\bar{1}00]$ are determined to be of the $\mathbf{b}=1/3[11\bar{2}0]$ type. These types of dislocations are reported to be mainly observed defects in GaN grown on various subtrates.[10,11,13–15]

The solid line in Fig. 1(c) is the projection of the $(1\bar{1}01)$ plane, which is drawn from the right edge of the mask. For the region above $\sim 2\ \mu$m from the interface, it can be seen that once the threading dislocations intersect the $(1\bar{1}01)$ plane, almost all of them change direction and become parallel to the $[1\bar{1}00]$ direction. These dislocations thread through the lateral-growth part of GaN. Thus, the density of the threading dislocations is effectively reduced. For the lower region, most of the dislocations running parallel to the $[1\bar{1}00]$ direction in the lateral-growth part of the pyramid are not generated by the bending upon crossing the $(1\bar{1}00)$ plane (the solid line), as can be seen in Fig. 1(c). They originate from dislocations parallel to the $[1\bar{1}00]$ direction in the window region, which are generated by coalescence of the nitride islands. These dislocations are either of the $\mathbf{b}=1/3[11\bar{2}0]$ type or the $\mathbf{b}=1/3[11\bar{2}3]$ type.

Now we consider the interface region. First, the composition of AlGaN was determined by EDX. The measurements were performed at several points approximately every 0.5 μm along the interface in the window region, since the contrast between AlGaN and GaN was not distinct. The results indicate that the AlN mole fraction varied due to the island-like structure, but the maximum value was found to be 0.1–0.12, which is consistent with the estimated value (0.09). In this connection, Hirono *et al.*[16] reported that an AlN-rich region is generated at the interface for the MOVPE growth of AlGaN on 6H-SiC(0001) substrates. Our result indicates that such segregation does not occur for the AlGaN growth on Si.

Next, the interface was characterized by high resolution TEM (HRTEM). It was found that an amorphous layer with an average thickness of approximately 3 nm is formed between the nitride and Si in the window region, as shown in Fig. 2. The amorphous layer is wavy and the thickness varies along the interface. By close inspection of the HRTEM image, Si particles are seen in some places in the amorphous layer, as determined by the fringe spacing. EDX measurements did not reveal any foreign elements in the interface region. Thus, we deduce that the interfacial amorphous layer is silicon nitride. It should be noted that in spite of the presence of the amorphous layer, there is a certain orientation relationship between the nitride and Si, as mentioned earlier.

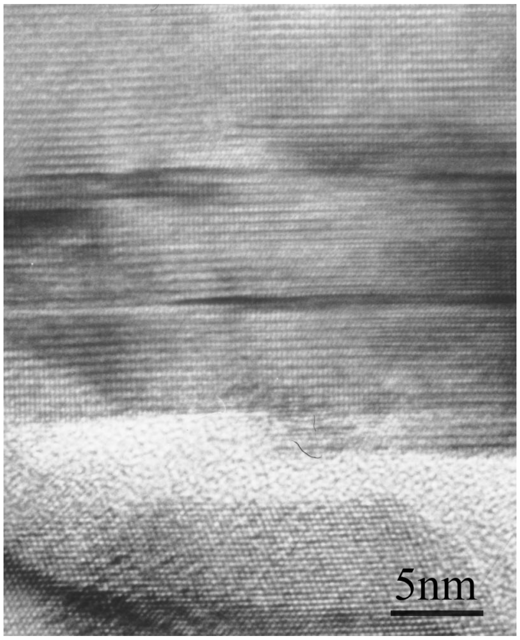

FIG. 2. High resolution TEM image of the interface between the nitride and Si.

This suggests that the amorphous layer is generated during the growth of the nitride. Under the condition of high growth temperature and high stress induced by the lattice mismatch between the nitride and Si, the mutual diffusion of N and Si and subsequent formation of the amorphous layer may occur. However, more work is required to understand the formation of the interfacial amorphous layer. Basal stacking faults are also seen near the interface in the window region. The presence of such faults is reported for sapphire[11,14,15] and 6H-SiC[17] substrates close to the interface.

Figure 3 is a HRTEM image of the interface in the mask region. A small particle of approximately 15 nm in size can be identified on the mask by the difference of the fringe pattern in the center region of the micrograph. Such interfacial particles were frequently observed on the mask. Fourier analysis of the Moiré fringe observed in the particle indicated the existence of planes with an interplanar distance equal to that of (0001) planes of the nitride, forming an angle of approximately 40° with the (0001) planes of the bulk of the nitride. Since the difference of the lattice parameter between GaN and $Al_{0.09}Ga_{0.91}N$ is small compared with the accuracy of the method, it was not possible to distinguish between GaN and $Al_{0.09}Ga_{0.91}N$. Nitride particles may remain on the mask during AlGaN and/or GaN growth and they may be buried upon the lateral growth of GaN.

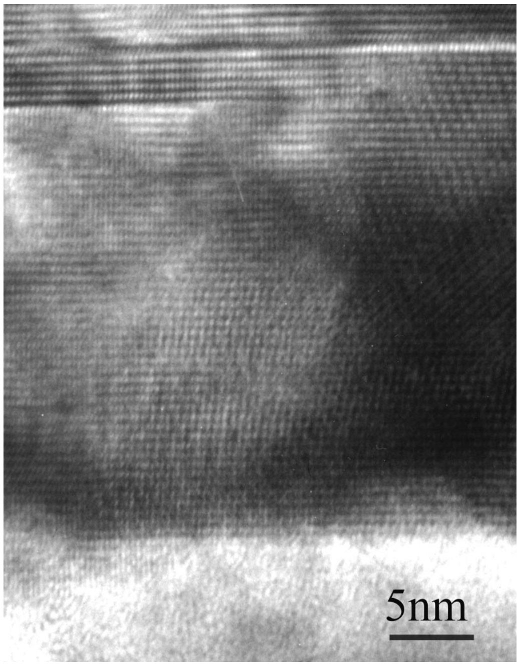

FIG. 3. High resolution TEM image of the interface between the nitride and the SiO_2 mask.

In summary, we have carried out TEM characterization of a GaN pyramid grown on (111)Si by MOVPE using AlGaN as an intermediate layer. The density of threading dislocations was very high near the interface region in the window, but it decreased gradually with increasing distance from the interface. The reduction of the threading dislocation density is mainly due to the dislocation reaction for the first ~2 μm region from the interface, and for the upper region it is mainly due to bending of threading dislocations. Dislocations parallel to the interface were dominantly observed in the lateral-growth part. An amorphous layer was formed at the interface in the window region, which is considered to be introduced during the growth of the nitride. Nitride particles were observed at the interface in the mask region.

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