Structural characterization of GaN laterally overgrown on a (111)Si substrate

Shigeyasu Tanaka,[a] Yoshio Honda, and Nobuhiko Sawaki
Department of Electronics, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan

Michio Hibino
Center for Integrated Research in Science and Engineering, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan

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Using transmission electron microscopy, we have characterized defect structures in laterally overgrown GaN crystals, grown directly on SiO_2 stripe-patterned (111)Si substrates by metalorganic vapor phase epitaxy using AlGaN as an intermediate layer. The width and the period of the stripe windows were nominally 1 and 2 μm, respectively. The average threading dislocation density for a completely coalesced 2-μm-thick GaN crystal obtained on the [11$\bar{2}$]-oriented stripe-patterned substrate was $\sim 2\times 10^9$ cm^{-2}. The reduction in threading dislocation density is a consequence of the lateral growth and dislocation reactions at the coalesced front of the mask. On the other hand, valleys and pits tend to remain on the mask during the growth on the [1$\bar{1}$0]-oriented stripe-patterned substrate. Cracks were present in both crystals.
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There has been considerable interest in the growth of GaN on Si substrates because of its low cost, large size, and the potential for the integration of GaN-based devices with Si-based ones.[1–3] However, the large difference in the lattice constant, the thermal expansion coefficient, and the surface chemical make high-quality epitaxy of GaN on Si difficult. These difficulties are alleviated by the use of a thin buffer layer such as AlN.[4] However, even with such a buffer layer, GaN crystals on Si contain many crystalline defects and cracks.[5] The threading dislocation densities are as high as 10^{10} cm^{-2}.

Recently, the epitaxial lateral overgrowth (ELO) technique, which was proven to be extremely effective in reducing the threading dislocation density in laterally overgrown GaN crystals on sapphire[6–11] and 6H-SiC[10,12,13] substrates, has been applied for the growth of GaN on Si. Kung *et al.*[14] obtained an ELO–GaN crystal on a GaN template previously grown on a (111)Si substrate. Also, Linthicum *et al.*[12] used a GaN seed layer on an AlN/3C-SiC/(111)Si structure and obtained a coalesced single-crystal layer of GaN. These growth processes are performed in two steps. On the other hand, Kawaguchi *et al.*[15] demonstrated a single growth step process for a selective area growth of GaN directly on a dot-patterned Si substrate. They used AlGaN grown at a high growth temperature as a nucleation layer in metalorganic vapor phase epitaxy (MOVPE). Subsequently, Honda *et al.*[16] used this technique for the growth of ELO–GaN crystals on stripe-patterned Si substrates. They characterized the ELO–GaN crystals by scanning electron microscopy, photoluminescence, and x-ray diffraction, and confirmed the improved properties of the ELO–GaN crystals. In this study, the microstructure of ELO–GaN crystals grown directly on Si is characterized by transmission electron microscopy (TEM).

[a]Electronic mail: tanaka@nuee.nagoya-u.ac.jp

ELO–GaN crystals were grown by atmospheric-pressure MOVPE, using AlGaN as an intermediate layer, on (111) Si substrates with a stripe-patterned structure of a silicon dioxide (SiO_2) mask. After the deposition of the SiO_2 film by radio-frequency sputtering on the substrate, stripe windows were patterned by the conventional photolithographic method and wet chemical etching. The width and the period of the stripe windows were nominally 1 and 2 μm, respectively. The stripe was oriented parallel to the [1$\bar{1}$0] or the [11$\bar{2}$] axis of silicon. Trimethylgallium, trimethylaluminum, and ammonia were used as source gases of Ga, Al, and N, respectively. The AlGaN intermediate layer was grown on the stripe-patterned Si substrate at 1100–1150 °C for 4 min, and then GaN was grown at 1050–1100 °C for 60 min. Both cross-sectional and plan-view TEM observations were carried out. Cross-sectional TEM samples for observation along the SiO_2 stripes were prepared using conventional procedures involving the sequence of grinding, polishing, and Ar-ion milling. Plan-view TEM samples were thinned from the substrate side in the same manner. To observe the near-interface region by plan-view TEM, the plan-view TEM sample was also thinned from the surface. The TEM observations were carried out using electron microscopes: Hitachi H-8000 operated at 200 kV or Hitachi H-1250ST operated at 1000 kV.

In the single growth step process, the growth of GaN was initiated by nucleation on AlGaN grains in the window region.[15] In the growth of AlGaN, grains of AlGaN are also present on the mask, but they do not function as a nucleation center for subsequent GaN growth if proper growth conditions are met.[17,18] By continuing the growth, GaN stripes bounded on the edges by inclined sidewalls are developed.[18] Then, adjacent GaN stripes laterally overgrown on the mask merge to form a continuous layer. ELO–GaN crystals grown in this way have a wurtzite structure and the standard orien-

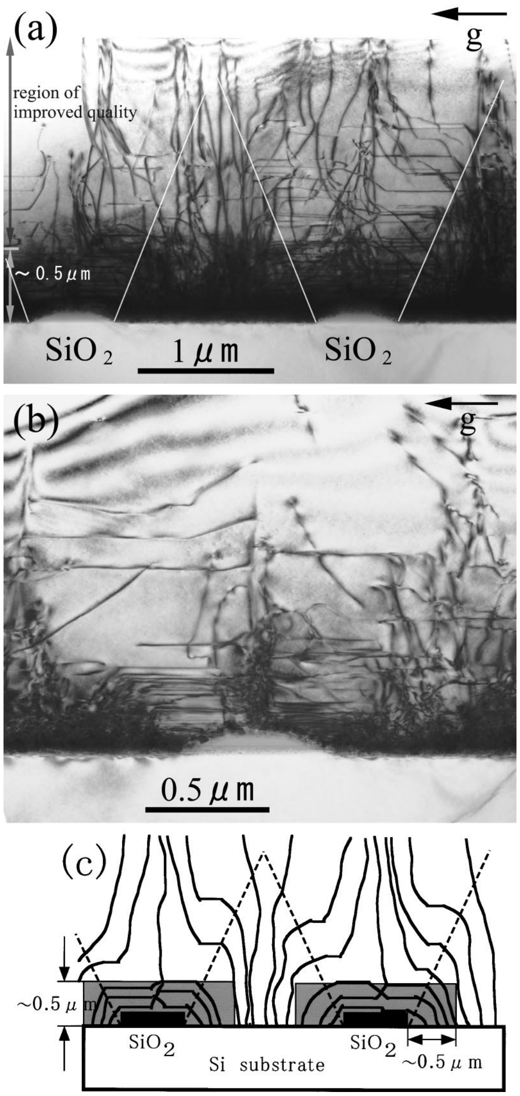

FIG. 1. (a) Cross-sectional TEM image of an ELO–GaN structure on the [112]-oriented stripe-patterned Si substrate. $\mathbf{g}=11\bar{2}0$. The white lines indicate the sidewall of GaN stripes developed during the growth. (b) Vicinity of the mask region. (c) Full view of the cross section of ELO–GaN. The heavy solid lines and the doted lines represent the dislocations and sidewalls of GaN stripes developed during the growth, respectively.

tation relationship with Si substrates as follows: $[11\bar{2}0]$GaN//[110]Si and [0001]GaN//[111]Si.

Figure 1(a) shows a cross-sectional TEM image of a typical ELO structure on a Si substrate with the [112]-oriented stripe pattern. The mask was somewhat overetched during the photolithographic process. Completely coalesced 2-μm-thick GaN crystals were obtained following the 60 min growth. For the [112]-oriented stripe-patterned substrate, each sidewall of the GaN stripes developed during the growth consists predominantly of two $\{1\bar{1}01\}$ facets. Thus, the growth front on the mask is zigzag-shaped along the stripe. Adjacent GaN stripes with such a sidewall rapidly merge into a planar structure over the mask, as shown in Fig. 1(a).

The main observed defects in Fig. 1(a) are threading dislocations generated at the interface in the window region. They initially propagate vertically along the growth direction until they meet the sidewall along the growth window edge. Once these threading dislocations intersect the facet, almost

FIG. 2. Plan-view TEM image of a near-interface region of an ELO–GaN structure on the [112]-oriented stripe-patterned Si substrate. Both window and mask regions are imaged. $\mathbf{g}=11\bar{2}0$.

all of them change direction and become parallel to the interface. This is due to the change in the growth mode from vertical to lateral. Most of the dislocations reaching the mask region are bent upward again over the mask, indicating that the coalesced region is buried by the vertical growth. The full view of the picture including the entire growth window and the mask is depicted schematically in Fig. 1(c).

The threading dislocations are mostly mixed type with a Burgers vector $\mathbf{b}=1/3\langle 11\bar{2}3\rangle$ or edge type with a Burgers vector $\mathbf{b}=1/3\langle 11\bar{2}0\rangle$, as determined by contrast analysis. Due to the bending of the threading dislocations, the emergence points at the surface eventually become randomly distributed over the entire surface. The average density of the threading dislocations is estimated to be $\sim 2\times 10^{9}$ cm^{-2} at the surface. This value is much smaller than that ($\sim 1\times 10^{10}$ cm^{-2}) obtained for bulk GaN crystals grown directly on Si with the same system used in these studies.

Figure 1(b) shows a cross-sectional TEM image of the vicinity of the mask on the [112]-oriented stripe-patterned substrate. A region containing many defects is observed in the center part of the mask. This region extends ~0.5 μm from the interface, and is obviously generated by the gathering of many laterally propagating dislocations in a small area of the coalesced front. These regions are indicated by hatching in Fig. 1(c). The laterally propagating dislocations originated from threading dislocations created within a ~0.5 μm region from the edge of the mask. It should be noted that few dislocations within this region extend to the upper part, indicating that dislocation reactions take place and thus most dislocations are confined to this region. Thus, above ~0.5 μm thickness, the quality of the film is improved [Fig. 1(a)].

The threading dislocation density in the near-interface region in the window is $\sim 1\times 10^{10}$ cm^{-2}, as estimated using plan-view TEM images such as those shown in Fig. 2. The reduction of the threading dislocation density is greater than the value expected simply from the ratio of the window area to the total area ($\sim 7\times 10^{9}$ cm^{-2}). Here, we used 1.4 μm for the width of the stripe window. Further reduction in the threading dislocation density is due to the dislocation reac-

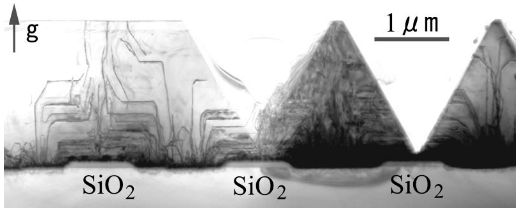

FIG. 3. Cross-sectional TEM image of an ELO–GaN structure on the $[1\bar{1}0]$-oriented stripe-patterned Si substrate. The blurred image of the center triangle is due to local bending. Glue used in the TEM sample preparation process accidentally remained in the valley or pit region. **g**=0002.

tion at the center region of the mask, as mentioned earlier. Since most of the threading dislocations created within a ~0.5 μm region from the edge of the mask do not extend to the surface, the mask may be effectively widened and thus the width of the stripe window may be effectively made narrow. The threading dislocation density estimated using the effective width of the stripe windows (0.4 μm) is consistent with the threading dislocation density at the surface.

According to the earlier results, the threading dislocation density is expected to be further reduced when a window width of less than ~1 μm is used. On the other hand, the mask width should be comparable to the width of the window in order to obtain a coalesced crystal within a realistic growth time. The surface of the ELO–GaN crystals on Si with the growth time beyond 60 min tends to be rough.[16] However, the period of the stripe window should be long, because residual threading dislocations leak from the coalesced region. Considering these factors, ~1 μm wide stripe windows with a periodicity of ~2 μm may be suitable for reducing the threading dislocation density. However, the threading dislocation density is still high compared to that on sapphire and 6H-SiC. In addition, cracks are present in the ELO–GaN crystals, as described later. Research to improve the crystalline quality is ongoing.

Next, an ELO–GaN crystal on the Si substrate with the [110]-oriented stripe pattern was examined. For the growth on the [110]-oriented stripe-patterned substrate, each sidewall of the GaN stripes consists of one {1101} facet. Since the {1101} facets are stable, and growth directly on these planes is difficult, valleys and pits tend to remain on the mask after the adjacent growth fronts meet following a 60 min growth, as can be observed in Fig. 3. In regions where the coalescence is complete and buried structures are obtained, the morphology of the threading dislocations is similar to that for ELO–GaN on the [112]-oriented stripe-patterned substrate. However, the majority of the threading dislocations are mainly located on the mask, because {1101} facets are sufficiently developed before the coalescence and most of the threading dislocations are gathered in the mask region.

Another defect in ELO–GaN structures on the Si substrate is cracks along the {1100} planes. Cracks occur due to large tensile stress while cooling to room temperature. The formation of cracks was similar for both stripe directions. The average distance between cracks is ~100 μm, which is comparable to the reported value[5] for bulk GaN crystals on Si.

In summary, we have carried out TEM characterization of ELO–GaN crystals grown on (111) Si by MOVPE, using AlGaN as an intermediate layer. On the [112]-oriented stripe-patterned substrate, the completely coalesced 2-μm-thick GaN crystal with an average threading dislocation density of $\sim 2\times10^9$ cm^{-2} was obtained following a 60 min growth. The reduced threading dislocation density is a consequence of the lateral growth and dislocation reactions. On the other hand, valleys and pits tend to remain on the mask during the growth on the [110]-oriented stripe-patterned substrate. Cracks are present in both types of ELO–GaN crystals.

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