A MICROSCOPIC EVALUATION OF THE SURFACE STRUCTURE OF OMVPE DEPOSITED α-GaN EPILAYERS

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ABSTRACT

The surface structure of organometallic vapor phase epitaxy (OMVPE) grown α-GaN films was investigated using optical and scanning force microscopy (SFM). Optical microscopy shows that the surface is decorated with several different types of faceted features that have lateral dimensions of 10 to 75 μm and occur with a density of approximately $10^4/cm^2$. SFM images show that on the flat regions of the surface, single diatomic layer steps, 2.6 Å high, are straight, evenly spaced (at 500 to 1500 Å intervals), and oriented along $\langle 10\bar{1}0 \rangle$ directions. The SFM images also show that the regular step patterns are often interrupted by faceted growth hillocks, 0.8 to 5 μm in diameter and 120 to 400 Å high, that occur with a density of $10^6/cm^2$. An open-core screw dislocation with a Burgers vector of 5.2 Å occurs at the center of each hillock and is a source for spiral steps. Other dislocations are also observed to intersect the flat regions of the surface and create a step, but these have smaller Burgers vectors, do not form spirals, and do not have open cores. Based on these observations, we conclude that thick OMVPE GaN films grow by a combination of the layer-by-layer and spiral growth mechanisms.

INTRODUCTION

Gallium nitride and its related alloys (AlGaN and InGaN) are important wide band-gap semiconductors that have potential applications in both short wavelength optoelectronic and high power/high frequency devices [1]. The most widely accepted technique for the deposition of nitride films and device structures, which was proposed by Amano [2,3] and Akasaki [4], uses sapphire substrates buffered by thin layers of AlN or GaN deposited at low temperature. However, films grown by nominally the same method can have very different microstructures and, accordingly, different properties. For example, the dominant defects in the films considered here are edge dislocations arranged in patterns that define low angle grain boundaries separating almost "dislocation free" grains [5]. The grain structure is columnar and, therefore, largely determined during the nucleation stage of growth. The objective of the work described in this report was to characterize the structure of the film growth surface in order to gain insight into the growth mechanism and its relationship to the through-thickness microstructure.

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EXPERIMENTAL

The 2.8 μm thick α-GaN epilayers described here have an (0001) orientation and were grown at 1040 °C on α-Al_2O_3(0001) substrates in an inductively heated, water cooled, vertical organometallic vapor phase epitaxy (OMVPE) reactor. [6] An AlN buffer layer was first deposited at 450-500 °C using 1.5 μmol/min triethylaluminum, 2.5 standard liters per minute (SLM) NH_3, and 3.5 SLM H_2 flows. After annealing in 2.5 SLM NH_3 and 3.5 SLM H_2 for 10 minutes at 1025 °C, GaN was grown using 49 μmol/min trimethylgallium (TMGa), 1.75 SLM NH_3, and 3.5 SLM H_2. The resulting growth rate was approximately 2.0 μm/hr. The as-grown films were examined in air with a traditional metallographic microscope and with a Park Scientific Instruments scanning force microscope (SFM). The 5 μm scanner was operated at 2 Hz. All images were acquired in the constant force mode using 3 to 12 nN of contact force.

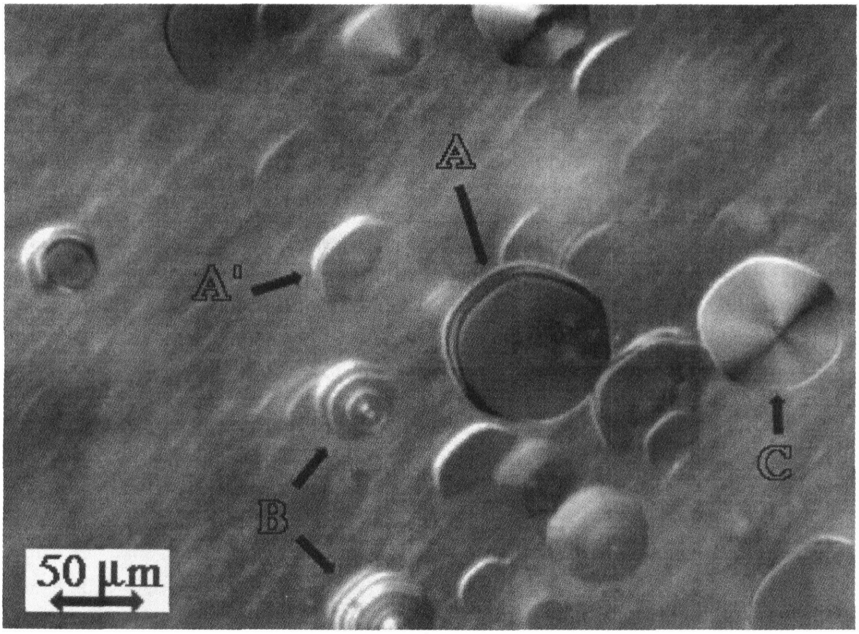

Figure 1. Optical micrograph of the growth surface of an OMVPE grown α-GaN epilayar.

RESULTS

When viewed under an optical microscope, a number of different topographic features were observed on the surface of the GaN epilayer. We classify all of the observed features into one of three groups: flat facets (labeled A and A' in Fig. 1), multifaceted hexagons (labeled B in Fig. 1), and cones (labeled C in Fig. 1). The average size and density of these features was determined by completely mapping a 2.02 mm x 2.25 mm area of the surface at 100X magnification. In this region, the observed density of flat facets is approximately 1.2 x $10^4/cm^2$. The image in Fig. 1 includes both the island-facets (65 to 75 μm in diameter) that are completely bounded by visible steps with prismatic orientations (labeled A in Fig. 1), as well as the more numerous ledge-facets (10 μm and larger) that are only partially bounded by visible steps (labeled A' in Fig. 1). The multifaceted hexagons have lateral dimensions of 30 to 45 μm and a density of 8 x $10^2/cm^2$. Most of these features have three concentric hexagonal steps separated by 6 to 8 μm. The density of the cone shaped features is 5 x $10^2/cm^2$ and they all are 65 to 75 μm in diameter. Some of the cones appear to come to a point while others have flat tops.

To the optical microscope, the surfaces of the island-facets appear flat. When the facets are imaged with the SFM, however, regular patterns of single layer steps and hexagonally faceted growth hillocks observed on the optically flat surface. The hillocks, such as the ones shown in Fig. 2, are composed of one to five concentric layers and are configured randomly; some of the hillocks have grown on top of each other, while others are separated by distances of more than 20 μm. The smaller hillocks are about 0.8 μm in diameter and 120 Å high, while the larger ones are more than 5 μm at the base and up to 400 Å high. Based on the analysis of one large, flat facet, we conclude that the hillock density is $1 \times 10^6/cm^2$.

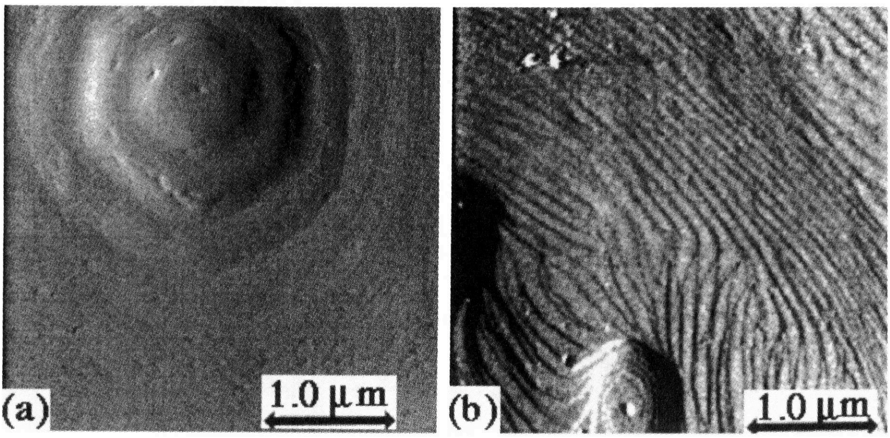

Figure 2. Growth hillocks on the surface of a flat facet on the GaN epilayer. These SFM images were recorded based on the difference between the setpoint deflection and the actual deflection of the probe so that contrast corresponds to changes in the topography. This allows a large dynamic range of height data to be viewed simultaneously. (a) shows one of the larger hillocks and (b) shows a smaller one in the lower portion of the image and single layer steps elsewhere.

Patterns of single layer steps are found between the hillocks. The steps are measured to be 2.8 ± 0.6 Å high. Although there is certainly some error introduced by the presence of the surface contamination layer, this measurement is consistent with the expected dimension of a single diatomic layer of GaN which is one half the length of the c-lattice parameter (2.6 Å). Far from the hillocks (see the upper right hand corner of Fig. 2b), the steps are straight and spaced at regular intervals of 500 to 1500 Å. However, in regions near the hillocks, the direction of step propagation is altered, as illustrated in Fig. 2b. Also, on the left hand side of the hillock, the steps spiral downward and remain evenly spaced. On the right hand side, however, they are bunched. This pattern is typical of all our observations.

We have observed two types of surface/dislocation intersections that we distinguish based on the magnitude of the Burgers vector normal to the surface and the effect that they have on the progress of growing steps. The first type is observed in the relatively flat areas of the surface between the hillocks where single steps are separated by approximately 1000 Å. For example, in the topographic SFM image in Fig. 3a, many surface steps (dark contrast) are clearly visible. Because some of these steps terminate in the middle of a terrace, a Burgers circuit around such a position indicates that a dislocation line must intersect the surface and that the component of the Burgers vector normal to the surface plane is equivalent to one half the length of the c-lattice parameter (2.6 Å). Therefore, the dislocation has at least partial screw character. Two such defects with opposite sign are indicated by arrows in Fig. 3a. By counting such defects in this and similar images, we find that their density varies greatly, but falls in the range 5 x10^6/cm^2 to 1 x10^8/cm^2.

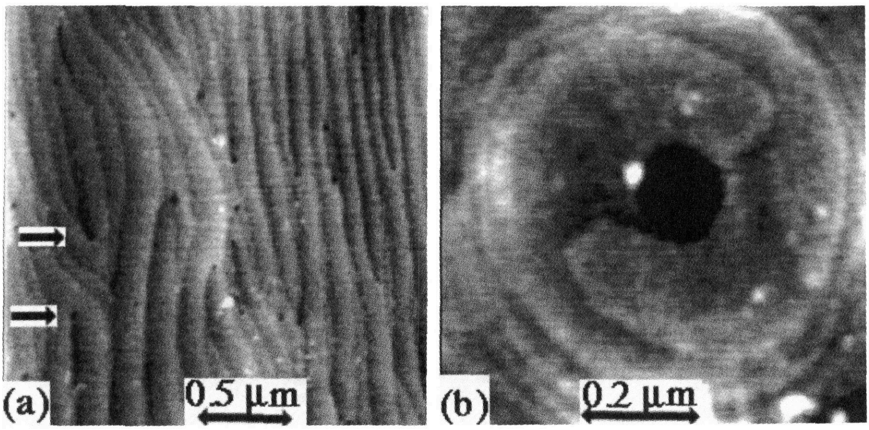

Figure 3. Topographic SFM images showing different types of surface/dislocation intersections. (a) was recorded on a relatively flat region. The vertical lines of contrast correspond to single layer steps. Where the steps end (positions indicated by the arrows), dislocations intersect the surface. (b) was recorded on top of a faceted hillock similar to the ones shown in Fig. 2a. The black spot in the center is the crater formed by the nanopipe that intersects the surface at this point. The spiral steps indicate that there is a screw dislocation at this position.

The second type of surface/dislocation intersection is found at the center of each of the faceted hillocks. SFM images recorded at the tops of these features (see Fig. 3b) illustrate that in the center of each one, there is a hole. Such images also show that a pair of spiral steps, each 3.1± 0.8 Å high, originates at this hole. This measurement is consistent with the expected dimension of a single diatomic layer of GaN which is one half the length of the c-lattice parameter (2.6 Å). Thus, the two steps form an additional complete GaN unit cell. The origin of this extra step at the center of the hillock indicates that there is a screw dislocation in the center of the hole with a Burgers vector (**b** = 1/3[0003]) equal in length to the c lattice parameter (5.2 Å). Numerous hillocks were examined with different probe tips and similar observations were made; each has a hole with an approximately 600 Å radius at the center. In one case, four single steps, each 1/2 c high, emerged from a single hole with a larger radius (approximately 925 Å). This corresponds to a "giant" dislocation with Burgers vector **b**=2/3[0003]. After spiraling away from the hole on the flat top of the hillock, the steps bunch together and become too close to be individually resolved. Similar features were observed on the flat facets and at the centers of the multifaceted hexagons.

DISCUSSION

The through thickness microstructure of these same films has been studied by transmission electron microscopy (TEM) and described in a previous report [5]. The film microstructure is dominated by submicron grains with relative misorientations of less than 3 °. The boundaries between adjacent grains are defined by arrays of edge dislocations, aligned along the [0001] growth direction and with Burgers vectors of $1/3\langle 11\bar{2}0\rangle$. There is no apparent relationship between this bulk microstructure and the observed surface microstructure. First, the faceted features observed in the optical microscope are all far larger than the observed submicron grain size of the columnar film. Furthermore, when flat areas with micron-scale dimensions were examined in detail with SFM, trains of parallel steps are present which presumably flow uninterrupted over the positions where the edge dislocation arrays that define the low angle boundaries intersect. Therefore, we conclude that the interaction between the steps and the threading edge dislocations is relatively weak and does not affect the progress of growing steps.

On the other hand, the dislocations of screw character that penetrate the surface serve as sources of steps. Those defects with Burgers vectors that are equal to the length of the GaN unit cell can form spirals and add a new layer of crystal to the film on each rotation. Considering the fact that these defects are found at the centers of the hillocks, we assume that these structures are formed by a spiral growth mechanism. The different sizes of these growth mounds suggest that they become active at different times during the growth of the crystal. Those dislocations with smaller Burgers vectors, which occur at the sources of straight steps on the flat regions of the crystal, are unable to create hillocks. Because the Burgers vector parallel to the c-axis is only one half the length of the lattice repeat distance, a stacking fault would be created on every rotation of the spiral. Thus, the other steps must grow over these defects in order for the crystal growth to continue. While steps show some curvature near these defects, they do not appear to act as strong pinning sites.

The voids that appear at the centers of spiral screw dislocations are known as nanopipes. Previously described TEM studies of these films concluded that these pipes penetrate the entire epilayer and occur with a density of 10^5 to $10^7/cm^2$ [7,8]. The stability of such defects was first demonstrated by Frank [9], who argued that a state of local equilibrium could be achieved by balancing the elastic energy of the dislocation against the surface energy of the facets that bound the pipe. One of the predictions central to Frank's theory is that when the stored elastic energy of the dislocation is sufficiently large, the core will be empty and its radius will be proportional to the square of the Burgers vector. In a qualitative sense, our observations are consistent with these ideas. For example, Frank predicted that the crater formed where the pipe meets the free surface is larger than the pipe itself. TEM observations indicate that nanopipes have radii in the 35-500 Å range, while the surface craters observed in SFM images are larger [8]. Also, the radius (r) of the crater of the super screw dislocation (b = 10.4 Å) was larger (r = 925 Å) than that of the single dislocation (b = 5.2 Å and r = 600 Å), as expected. Finally, the dislocations with Burgers vectors smaller than the c-axis repeat distance that were observed on the flat areas of the surface do not have open cores. Since the elastic energy scales as the square of the Burgers vector and these dislocations have Burgers vectors that are 1/2 the length of those with open cores, they store one quarter of the elastic energy and are proportionally more stable.

We should also note, however, that there are two apparent inconsistencies between our observations and Frank's prediction. First, incorporating two known quantities, the observed Burgers vector (b = 5.2 Å) and the smallest observed radius (r = 35 Å), into Frank's formula, we extract a ratio of the surface free energy to the shear modulus which is equal to 0.01 Å. For most materials, however, this ratio is 0.25 Å. If one assumes that the shear modulus is as high as 400 GPa, the surface energy would be only 40 mJ/m^2, a physically unlikely value. The second inconsistency is that the radii of the holes should have a discrete distribution of sizes proportional to $(\mathbf{nb})^2$, where n is an integer ≥ 1. Instead, TEM observations suggest a more random distribution of sizes within the range of 35 to 500 Å [7].

CONCLUSION

By the time the GaN epilayer is 2.8 μm thick, growth occurs by both a layer-by-layer and a spiral mechanism. The underlying bulk microstructure of low angle grain boundaries is presumably fixed in the nucleation stage and does not seem to affect the growth of the film in later stages. While the surface has some large topographic features with a density of approximately 10^4/cm^2, it also has flat regions that are dominated by single diatomic layer steps. These steps are 2.6 Å high, straight, evenly spaced (at 500 to 1500 Å intervals), and oriented along $\langle 10\bar{1}0\rangle$ directions. These regular patterns are often interrupted by faceted growth hillocks, 0.8 to 5 μm in diameter and 120 to 400 Å high, that occur with a density of 10^6/cm^2. An open-core screw dislocation with a Burgers vector of 5.2 Å occurs at the center of each hillock and is a source for spiral steps. Other dislocations are also observed to intersect the flat regions of the surface and create steps, but these have smaller Burgers vectors, do not form spirals, and do not have open cores.

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