Open-core screw dislocations in GaN epilayers observed by scanning force microscopy and high-resolution transmission electron microscopy

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Structural investigations of organometallic vapor phase epitaxy grown α-GaN films using high-resolution transmission electron microscopy and scanning force microscopy have revealed the presence of tunnel-like defects with 35–500 Å radii that are aligned along the growth direction of the crystal and penetrate the entire epilayer. These defects, which are termed "nanopipes," terminate on the free surface of the film at the centers of hexagonal growth hillocks and form craters with 600–1000 Å radii. Either one or two pairs of monolayer-height spiral steps were observed to emerge from the surface craters which allowed us to conclude that nanopipes are the open cores of screw dislocations. The measured dimensions of the defects are compared to Frank's theory for the open-core dislocation. © *1995 American Institute of Physics.*

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] It is widely accepted that both the external efficiency and reliability of light emitting devices are sensitive to the type and density of extended defects in the material. Nitride films deposited on sapphire, which is poorly matched to GaN both in terms of lattice parameter and thermal expansion coefficient, typically exhibit dislocation densities in the 10^{10} cm^{-2} range.[2,3] Most of these dislocations are of pure edge type forming low angle "twist" boundaries. Recently, we have reported observations of another type of defect which could have a profound impact on characteristics of high voltage power devices.[4] These defects, referred to as nanopipes, are long, faceted empty pipes which thread through the entire thickness of the GaN epilayer. The radii of the pipes are in the 35–500 Å range and they appear to propagate along the *c* axis of the film. A similar defect is frequently observed in another wide band-gap semiconductor with a hexagonal crystal structure, namely, silicon carbide.[5] The present study, which combines high-resolution transmission electron microscopy (HRTEM) and scanning force microscopy (SFM), provides evidence that the nanopipes occur at the cores of screw dislocations. SFM images show that spiral steps emerge from the crater formed where the screw dislocations intersect the surface. These spiral steps create hexagonal growth hillocks which eventually lead to a nonplanar surface morphology. Although we believe that these observations can be understood in terms of Frank's theory for open-core dislocations, we note several quantitative discrepancies.[6]

The 2.8-μm-thick α-GaN epilayers described here 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.[7] 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 min 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/h. The as-grown films were examined in air with a Park Scientific Instruments SFM. The 5 μm scanner was operated at 2 Hz. All images were acquired in the constant force mode using 3–12 nN of contact force. TEM specimens were prepared by mechanical polishing, followed by Ar^+ sputtering at liquid-nitrogen temperature (both from the substrate side). TEM observations were made using a JEOL 4000EX-TEM, operated at 400 keV.

Examination of the film surfaces using an optical microscope at 750× magnification reveals hexagonal growth features, 10–100 μm in diameter, that are bounded by concentric sets of $\{1\bar{1}00\}$ steps that appear to form closed loops. Each of these features is capped by an optically flat (0001) surface. By positioning the SFM probe in the center of the flat top of such a feature, it was possible to image smaller hillocks such as the one shown in Fig. 1, that are also bounded by $\{1\bar{1}00\}$ steps along $\langle 11\bar{2}0\rangle$ and capped by a flat surface.The smaller hillocks are 1–5 μm in diameter, approximately 300 Å high, and are staged into layers that are each 100–200 Å high. Higher resolution SFM images, such as the one shown in Fig. 2, illustrate that there is a hole (the black spot) in the center of each hillock. The topographic profile recorded as the tip moved across the different layers of the hillock and into the hole in its center is shown in Fig. 1(c). Because the radius of the SFM probe is comparable in size to the radius of the hole, the profile does not reveal the true depth or three-dimensional shape of the hole.

The image in Fig. 2 shows that a pair of spiral steps, each 3.1±0.8 Å high, originates at this hole. Although there is certainly some error introduced by the presence of the

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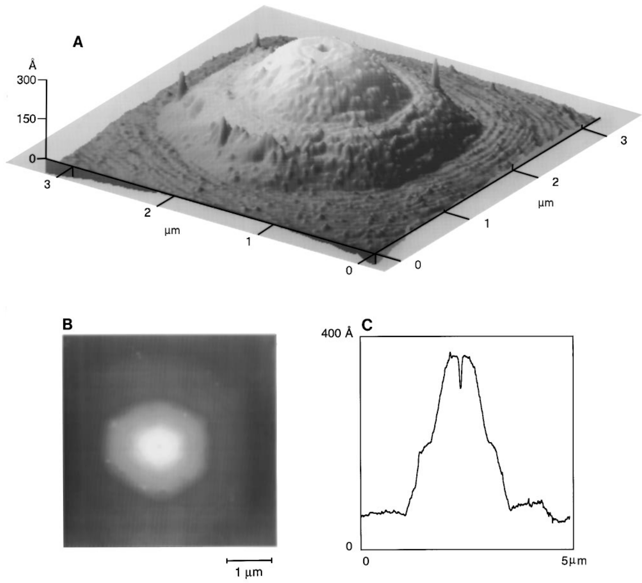

FIG. 1. SFM images taken from a typical growth hillock. (a) Three-dimensional view, showing that the smaller hillocks are staged into layers that are each 100–200 Å high. (b) Two-dimensional image, showing that the hillock is bounded by $\{1\bar{1}00\}$ steps along $\langle 11\bar{2}0\rangle$. (c) Line profile across the hillock center showing the central nanopipe.

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). 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 ($\mathbf{b}=1/3[0003]$) 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 the same hole which had a larger radius (approximately 925 Å). This corresponds to a "giant" dislocation with Burgers vector $\mathbf{b}=2/3[0003]$.

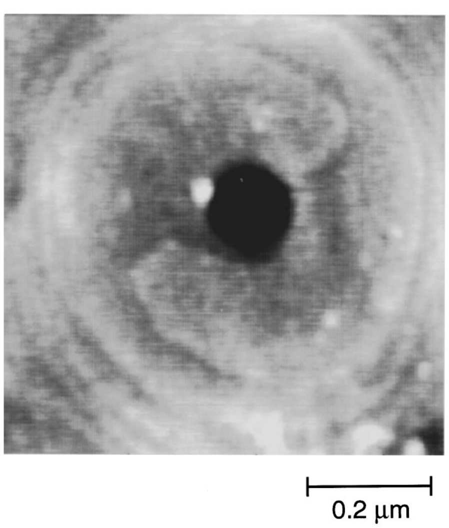

FIG. 2. High-resolution SFM image of the open-core screw dislocation at the hillock center. A pair of monolayer spiral steps (3.1±0.8 Å) originate from the open core and propagate outwards.

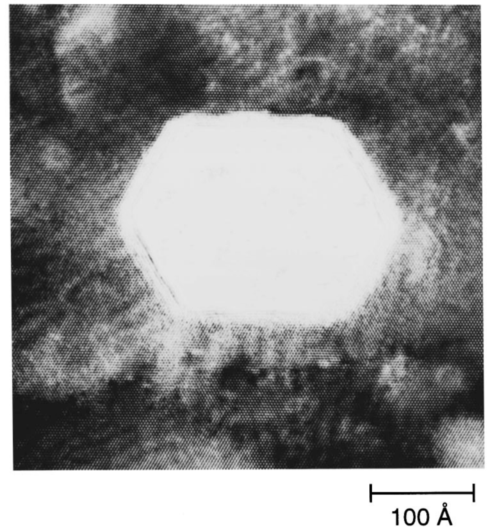

FIG. 3. Atomic resolution TEM image of a screw dislocation open core obtained with the [0001] zone axis. The internal surfaces of the open core are formed by six close-packed $\{1\bar{1}00\}$ prism planes (lattice spacing of 2.76 Å).

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.

In an earlier study, long hollow pipes that were similar in size, shape, and density were also observed using conventional TEM.[4] The defects are stable under electron beam illumination and the atomic scale structure can be determined from HRTEM images taken in the [0001] orientation (see Fig. 3). The internal surfaces of the open core are formed by six close-packed $\{1\bar{1}00\}$ prism planes (lattice spacing of 2.76 Å) that have the same morphology as the growth hillock. Because this image shows a projection of the atomic structure parallel to the Burgers vector, a closed circuit drawn around the open core shows no net atomic displacement. This was also the case for several other nanopipes examined in detail by HRTEM. Considering the fact that edge dislocations parallel to the *c* axis outnumber screw dislocations by orders of magnitude and that no nanopipes with edge character were observed, it seems unlikely that edge dislocations have open cores.[3] This can be explained by the fact that the Burgers vectors of screw dislocations ($\mathbf{b}=1/3[0003]$) are 62% larger than those of edge dislocations ($\mathbf{b}=1/3\langle 11\bar{2}0\rangle$) and, therefore, have a much larger elastic energy.

Our microscopic observations indicate that the density of the nanopipes is 10^5–$10^7/\mathrm{cm}^2$. The large error in the determination stems from the small size and low density of the defect; the observations must be carried out in a high magnification mode and only relatively small areas can be searched. The radii of the nanopipes measured by TEM vary from 35 to 500 Å. The apparent discrepancy between the nanopipe sizes determined from the SFM and TEM images is resolved by recognizing that the SFM image actually measures a crater on the growth surface that is always wider than the nanopipe itself. The crater can be easily observed in plan-view TEM specimens which preserve the GaN growth surface[4] and is a predicted structural component of an open-core dislocation.[6]

We were unable to identify nanopipes in cross-sectional TEM. One reason for this is that they have a low density and would occur very rarely in a random thin cross-section specimen. A second complication is that they could only be distinguished from a regular dislocation if the sample thickness were comparable in size to the nanopipe diameter. Without cross-sectional observations, it is difficult to determine whether or not the tubes penetrate the entire film, from the growth surface to the substrate. We addressed this issue by making a series of plan-view observations from a single TEM specimen that was progressively ion milled from its growth surface. This procedure permits examination of regions at various distances (although not exactly known) from the free surface. Because empty pipes can always be observed in the sample, we conclude that the defects penetrate through the thickness of the GaN film.

Scanning probe microscopy has been used to observe spiral growth steps with single unit cell step heights in a number of other epilayers grown from vapor.[8,9] However, the previously reported growth features did not have hollow central cores. The defects we observe in GaN appear to be more closely related to, but two orders of magnitude smaller than the micropipe defect that occurs in SiC,[5,10] some flux grown garnets,[11] and a number of other materials. The SiC micropipes have received the greatest attention because they are known to be the defects that limit the breakdown voltage of high power devices.[12] The Burgers vector associated with the spiral steps in SiC is usually greater than 100 Å and the defect is, therefore, visible in conventional optical microscopes.

The theoretical stability of the empty core dislocation was first predicted by Frank,[6] 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. The relevant physical parameters that determine the proportionality are the surface energy and the shear modulus. When typical values are considered, a dislocation with a Burgers vector on the order of, or greater than, 10 Å will have a stable open core. While earlier observations of dislocations that have open cores have been consistent with this prediction, not all dislocations with Burgers vectors larger than 10 Å have open cores.

Qualitatively, our observations are consistent with Frank's theory. First, Frank predicted that the crater formed where the pipe meets the free surface is larger than the pipe itself, as observed. Second, the radius of the crater (r) of the super screw dislocation ($b=10.4$ Å) was larger ($r=925$ Å) than that of the single dislocation ($\mathbf{b}=5.2$ Å and $r=600$ Å). Quantitatively, however there are two apparent inconsistencies. First, incorporating two known quantities, the observed Burgers vector ($\mathbf{b}=5.2$ Å) and the smallest observed radius ($r=35$ Å), into Frank's formula,[6] 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 $(n\mathbf{b})^2$, where n is an integer $\geqslant 1$. Instead, TEM observations suggest a more random distribution of sizes within the range of 35–500 Å.

Although there are quantitative inconsistencies between Frank's theory and our observations, the qualitative agreement suggests that the theory is fundamentally valid and can account for nanopipe formation. Kinetic effects that depend on the deposition conditions and the thermal history of the sample might be the source of the quantitative discrepancies between our observations and the theory. Assuming that the hollow pipes will be a detriment rather than an asset to device properties, as they are in SiC, it will be necessary to determine the processing conditions in which the open cores become unstable and vanish, as Frank's theory seems to predict.

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