Nanomanipulation of ridges in few-layer epitaxial graphene grown on the carbon face of 4H-SiC

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\textbf{Abstract.} The atomic force microscope (AFM) is used to study the morphology of graphene grown on 4H-SiC\textsuperscript{(0001)}. A mesh-like network of ridges with high curvature is revealed that bound atomically flat, tile-like facets of few-layer graphene (FLG). To further study the structural properties of the ridge network, nanomanipulation experiments are performed using an AFM tip to deform the ridges in both the vertical and lateral directions. From these experiments, evidence is obtained that the ridges can be displaced in both the vertical and lateral directions. In some instances, ridges are found to return to their original shape after deformation. Cross-section transmission electron microscopy (TEM) studies show that the ridges are formed by the delamination of FLG from the SiC substrate.
1. Introduction

High-quality graphene, consisting of a single layer of carbon atoms arranged in a dense honeycomb lattice structure, has many unique electronic properties that are of considerable current interest [1, 2]. Various methods, including micro-mechanical cleavage, chemical exfoliation and epitaxial growth, have been proposed to prepare few-layer graphene (FLG) [3]–[5]. The epitaxial approach is promising, because it has the capability of producing wafer-scale graphene by simply heating a substrate that exhibits a tendency toward graphitization. Indeed, the preferential sublimation of Si atoms from a SiC substrate [4, 6, 7] offers an appealing way to grow FLG on wafer-size substrates, but the optimal growth conditions have not yet been identified.

High-quality graphene is required for advanced electronic applications and it is therefore not surprising that the structural characterization of FLG on SiC has been studied by a variety of surface science techniques, such as low-energy electron microscopy (LEEM) [8, 9], x-ray photoelectron spectroscopy (XPS) [10, 11], Raman spectroscopy [12, 13] and scanning probe microscopy [14, 15]. Previous work has shown that the graphitic material grown on the C face of 4H-SiC(0001) over a range of growth temperatures $\gtrsim 1475 \, ^\circ \text{C}$ produces a continuous layer of graphitization. In general, the graphite material that forms is atomically smooth over dimensions spanning a few microns. These smooth, tile-like facets of FLG are bounded by an interlocking mesh of ridge-like surface features [16]–[18]. The mesh-like network roughens the FLG to minimize the elastic strain energy of the thin graphitic layer [19]. Due to the high radius of curvature associated with the ridges, the likelihood of reactive sites that promote chemical oxidation increases. As a result, it seems possible that the quality of FLG decorated with ridge-like networks will degrade with time, especially if stored under ambient conditions.

There are few reports on the structural properties of these ridges. Previous work using high-resolution scanning tunneling microscopy (STM) has shown that flat regions of FLG tend to exhibit moiré patterns close to the ridges, indicating that ridge formation may produce a mosaic spread in graphene layer orientation [15]. Angular mismatches of a few degrees are
commonly inferred from the periodicity of the moiré patterns. Other studies have shown that the ridges are corrugated parallel to their length, indicating buckling of the graphene layers at the atomic scale [20]. Yet another study that focused on the characterization of the ridge network has provided quantitative estimates for the strain at any point along the ridge, conclusively demonstrating that stored elastic energy is inhomogeneously distributed in FLG layers [19].

To further investigate the structural and mechanical properties of the ridges in FLG, atomic force microscope (AFM) nanomanipulation techniques have been used to study the ridges in the graphitic layers that form at growth temperatures between 1525 and 1600 °C on the C face of 4H-SiC(0001). One of the major findings reported below is that the ridge network is flexible and can be compressed vertically and displaced laterally by the AFM tip.

2. Experimental techniques

2.1. Sample preparation

The SiC substrates used in this study are 75-mm-diameter 4H-SiC(0001) semi-insulating substrates obtained from CREE with a nominal cut-off angle of nearly 0°. The as-received SiC wafers were chemically polished by NovaSiC. After dicing the polished wafer into 1 cm × 1 cm samples, the individual SiC samples are solvent-cleaned and then etched in a piranha solution to remove hydrocarbon contamination. Before carbon growth, the samples are subjected to hydrogen etching at 1500 °C for 10 min to eliminate surface damage caused by wafer polishing in an Epigress VP508 SiC hot-wall chemical vapor deposition (CVD) reactor. After cooling below 700 °C, the chamber is pumped until the pressure is reduced to ~4 × 10⁻⁶ mbar. Subsequently, the temperature of the SiC is ramped to a pre-set growth temperature. Typically, the growth pressure is ~5 × 10⁻⁵ mbar. The temperatures reported in this study are measured using a Heitronics KT81R two-color rationing pyrometer (spectral bands 0.7 and 1.2 μm) with a calibration traceable to the melting temperature of Si at 1410 °C.

In this study, FLG samples were grown at a fixed growth temperature—ranging from 1525 to 1600 °C—that was held constant for 10 min. After FLG growth, the power to the radio frequency (RF) generator was switched off and the sample was allowed to cool to room temperature. The temperature during cooldown as a function of time \( t \) (in units of seconds) is well described by the equation \( T(t) = T_G e^{-\alpha t} \), where \( T_G \) is the growth temperature and \( \alpha \approx 8.2 \times 10^{-4} \text{s}^{-1} \).

From previous studies [19], the approximate thickness of FLG can be estimated by measuring the attenuation of the C-1s electrons from the SiC substrate as a function of tilt angle. This technique works well for sufficiently thin layers of FLG that do not completely attenuate the C-1s XPS signal and serves as a reliable guide for estimating the thickness of the FLG grown in the Epigress reactor. In what follows, we rely on this calibration to estimate the thickness of the FLG used in this study. A plot of the graphene thickness versus growth temperature is given in figure 1.

2.2. Atomic force microscopy (AFM)

A Nanotec Electronica AFM was used to image the graphene grown on SiC substrate. AFM images were obtained in an intermittent contact mode with a set-point amplitude reduction of 80%. The AFM studies were performed under ambient conditions. The \( z \)-axis of the AFM was calibrated against the step height of highly oriented pyrolytic graphite (HOPG).
Figure 1. Thickness, inferred from XPS measurements, of the graphene layer that forms on hydrogen-etched 4H-SiC(0001) in 10 min versus growth temperature. Data are from [19].

The x, y calibrations were performed using a Micromasch (model TGZ01) xyz calibration grating. WSxM software was used to both acquire and analyze the images [21]. Microcantilevers, purchased from Nanosensor (model SSS-NCL-10), with a nominal resonance frequency of 190 kHz and a nominal spring constant of 48 Nm$^{-1}$, were used throughout. In this study, both normal ($F_N$) and lateral ($F_L$) forces were measured. The photodiode normal sensitivity ($S_z$) and the microcantilever out-of-plane stiffness ($k_z$) were required for calibrating the normal force of the microcantilever. $S_z$ was obtained by measuring the slope of the force–displacement curve over a hard substrate such as Si wafer. In preliminary experiments, we relied on estimates of $k_z$ for the microcantilever provided by the manufacturer. These are nominal values and can be in error by a factor of 2. In more careful experiments, the actual value of $k_z$ for the microcantilever was estimated using the method described by Sader et al [22]. The Sader et al method requires a measurement of both the resonance frequency and the $Q$-factor of the microcantilever, quantities that can be accurately determined from a measurement of the frequency response of the microcantilever. In addition, the dimensions (length and width) provided by the manufacturer of the microcantilever are used to implement the Sader et al method. To estimate the lateral force, we assumed a value of $\mu = 0.03$ for the coefficient of friction for graphite, as is found in the literature [23, 24]. The calibration for the lateral force was then estimated by assuming $F_L = \mu F_N$ when the tip was moved across a flat surface.

Nanomanipulation was achieved using the positioning capability inherent in the Nanotec Electronica AFM system. The AFM lithographic capability in WSxM software [21] was used to precisely control the normal force applied to the substrate in the contact mode AFM as well as the path of the AFM tip. In this way, the tip could be directed to push a ridge with a controlled lateral force in a well-defined direction.

The nanomanipulation experiments proceeded as follows. A ridge in FLG was located using standard intermittent contact AFM techniques. From the resulting image, it was possible to use software to mark the beginning and ending points to define a contact mode scan. Therefore, a single nanomanipulation experiment consisted of a contact mode scan between two well-defined points with a specified normal force and scan speed. During the nanomanipulation of the ridge, the normal and lateral forces were recorded along the line joining the two points. After performing this maneuver, an intermittent contact AFM image was obtained to assess the effect of the nanomanipulation. Typically, a complete nanomanipulation experiment involved a sequence of such scans performed at increasing normal force.

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Figure 2. An intermittent contact AFM image of a typical region of FLG grown on hydrogen-etched 4H-SiC(0001) at 1600 °C for 10 min. From figure 1, we infer an FLG thickness of \( \sim 15 \) layers of graphene. The image was obtained under ambient conditions. Atomically flat regions of the FLG surface with \( \sim 2 \mu \text{m} \times \sim 2 \mu \text{m} \) size are bounded by an interconnected ridge network. The labels \( a_1, a_2 \) indicate a few of the primary ridges that are connected at ridge nodes labeled \( b_1, b_2 \). The primary ridge \( a_1 \) is 10–15 nm high. The label \( c \) denotes a secondary ridge network of lower height. A distinctive feature of secondary ridges is the eventual incorporation of the ridge into the flat, tile-like FLG surface.

2.3. Transmission electron microscopy (TEM)

An FLG sample grown at 1600 °C was prepared for cross-sectional TEM analysis with a focused ion beam (FIB) liftout method using an FEI Nova dual beam FIB/SEM equipped with a Klöcke nanomanipulator [25]. Protective layers of Pt/C were deposited locally, first with an electron beam to avoid surface damage, followed by a thicker layer deposited using an ion source. Primary milling was performed with the ion source operating at 30 kV, with a final cleaning at a lower voltage of 5 kV. TEM images were obtained with an FEI Titan 80-300 operating at 300 kV, equipped with a Gatan Image Filter with a 2 K CCD camera. Care was taken to limit the electron dose while imaging.

3. AFM results

3.1. The few-layer graphene (FLG) surface

A continuous layer of graphitic material forms at growth temperatures of 1475 °C and higher. The typical surface morphology that develops at 1600 °C is shown in the AFM image presented in figure 2. It is evident that the FLG surface exhibits a two-dimensional (2D) network of
interconnected one-dimensional (1D) ridges. The ridges are not of uniform height and contain a number of localized ridge nodes where individual ridges intersect. Overall, the FLG surface bears a resemblance to the surface of a piece of crumpled paper that is unfolded to a flat state. The flat facets have been extensively characterized in STM studies that show atoms arranged in a hexagonal network with a spacing of 0.245 nm [15].

As discussed previously [19], the ridges can be categorized into primary and secondary ridges. The labels \(a_1, a_2\) in figure 2 denote primary ridges, which tend to be interconnected with each other at ridge nodes, denoted by the letters \(b_1\) and \(b_2\). The ridge nodes tend to come in two types: tightly interconnected \((b_2)\) and loosely interconnected \((b_1)\). The height of the prominent ridge \(a_1\) varies between 15 and 20 nm. Secondary ridges \((c)\) of lesser height are also observed and often have the appearance of creases or pleats in the flat FLG facets. The secondary ridge \(c\) in figure 2 has a height of 2–10 nm. The distinction between a primary ridge and a secondary ridge is as follows: (i) the maximum height of a secondary ridge is typically less than the height of a primary ridge; (ii) a secondary ridge often appears to emanate from a primary ridge and forms a crease-like distortion on the surface of a flat, tile-like graphitic facet; and (iii) the height of a secondary ridge gradually decreases to 0 as it merges into a flat facet, while a primary ridge maintains a reasonably constant height between two ridge nodes. In parallel STM studies of identical SiC samples, we found evidence for moiré superlattices on the flat tile-like facets that were attributed to the relative rotation between two graphene layers [15]. The occurrence of a moiré superlattice seems to be in close proximity to a ridge network.

The ridges are interconnected, often diverging from a well-defined ridge node by forming a subtended angle, \(\Theta\). As described elsewhere, AFM imaging studies have revealed preferred values for the angle \(\Theta\) near 60° ± 10° and 120° ± 10°. Since these angles are close to high-symmetry directions in the graphene lattice, we infer that ridge formation tends to occur along high-symmetry directions in the FLG film.

### 3.2. Force versus tip displacement on a ridge

By positioning the tip over a ridge, it was possible to perform a normal force versus displacement experiment to probe the stiffness of the tip–ridge contact. The results of such an experiment are summarized in figure 3, which illustrates a zoomed-in secondary ridge that appears in the FLG grown at 1525°C. Figure 3(a) shows a magnified image of the secondary ridge and figure 3(b) shows a line profile perpendicular to the ridge axis. From the line profile, the height of the secondary ridge is \(\sim 8\) nm. The apparent width of the ridge in figure 3(b) is greatly distorted by tip dilation effects. Marked in figure 3(a) are two points \(P_1\) and \(P_2\) where normal force versus \(z\)-displacement data were carefully measured. The \(z\)-displacement of the piezo is the sum of the cantilever bending (spring constant \(k_c\)) and sample indentation (spring constant \(k_s\)). The effective stiffness of the contact \((k_{\text{eff}})\) is composed of two contributions in series: the cantilever stiffness and the tip–sample contact stiffness. The two springs add in series, so the effective spring constant is given by

\[
\frac{1}{k_{\text{eff}}} = \frac{1}{k_c} + \frac{1}{k_s}.
\]

The effective stiffness of a tip–substrate contact can be estimated from the slope of the normal force versus \(z\)-displacement data collected when the tip is in contact with the substrate. When the tip is positioned over the flat FLG substrate \((P_1)\), the normal force versus \(z\)-displacement data reveal the expected linear increase in force as a function of the \(z\)-displacement (data not...
Figure 3. A close-up AFM image of a secondary ridge found in FLG grown on hydrogen-etched 4H-SiC(0001) at 1525°C for 10 min. The height of the ridge is \(\sim 8\) nm, as shown in (b). From figure 1, we infer an FLG thickness of \(\sim 9\) layers of graphene. In (a), the straight line marks the location of the height profile given in (b). The points \(P_1, P_2\) label the positions where the force versus distance indentation data are obtained. In (c), the results of the force versus displacement indentation experiment at \(P_2\) are given. The data indicate an initially soft ridge with an effective spring constant of \(\sim 9.8\) N\(\text{nm}^{-1}\), which stiffens after a displacement of \(\sim 8\) nm. In (d), a schematic interpretation that shows a ridge deformation consistent with the data is shown.

Assuming that the substrate is very hard, the measured slope of these data gives an effective stiffness of \(k_{\text{FLG}} = 22.8\) nN\(\text{nm}^{-1}\), which is in good agreement with the calibrated value obtained using the Sader et al method. When the tip is positioned over the ridge at point \(P_2\) and the normal force versus \(z\)-displacement experiment is repeated, the data reveal the presence of two distinct slopes as shown in figure 3(c). Initially, the slope of the indentation is measured to have an effective spring constant \(k_{\text{eff}} = 9.8\) nN\(\text{nm}^{-1}\), indicating that the ridge is considerably softer than the flat substrate. This behavior persists for a \(z\)-displacement distance of \(\sim 8\) nm. Upon further indentation, the normal force versus \(z\)-displacement data reveal an abrupt change in slope, acquiring a value of \(k_2 = 22.8\) nN\(\text{nm}^{-1}\), a result already measured when the tip was at the point \(P_1\).

From equation (1), the secondary ridge stiffness of \(k_s = 17.2\) nN\(\text{nm}^{-1}\) is estimated. This behavior can be qualitatively understood if the ridge is assumed to consist of graphene layers that have delaminated from the substrate, as shown schematically in figure 3(d). Upon pushing on the ridge with the AFM tip, the delaminated FLG easily deforms until the tip encounters the substrate. Subsequent AFM scans revealed no permanent damage to the ridge.
Figure 4. A close-up AFM image of a secondary ridge node found in FLG grown on hydrogen-etched 4H-SiC(0001) at 1500 °C for 10 min. The measured ridge height is about 7 nm. From figure 1, we infer an FLG thickness of ~7 layers of graphene. In (a), an intermittent contact AFM image obtained under ambient conditions is shown. A fiducial line in the upper half of the image serves to locate the ridge node. The black arrow shows the direction of a contact mode scan performed during a nanomanipulation experiment. In (b), the resulting AFM image after nanomanipulation is shown. The ridge node is displaced by ~200 nm. The subtended ridge angle also increases by ~8°.

3.3. Permanent lateral ridge displacement

By positioning the tip next to a ridge, it was possible to explore the lateral deformation of a secondary ridge by performing a nanomanipulation experiment that sweeps the tip across the ridge. The results of a typical experiment are shown in figure 4, which shows a secondary ridge found in FLG graphene grown at 1500 °C. In figure 4(a), an intermittent contact AFM image of the ridge is shown. From this image, the ridge height is found to be ~7 nm. A black line in the upper half of the image has been added as a fiducial mark to accurately locate the position of the ridge node. After acquiring the image in figure 4(a), a nanomanipulation experiment was performed by sweeping the tip across the ridge in contact mode along a line indicated by the black arrow. Figure 4(b) shows the resulting intermittent contact AFM image. It is evident that the ridge node has been permanently displaced by ~100 nm relative to the black fiducial line. The angle θ subtending the ridge node has also opened up, changing from Θ_i = 128 ± 3° to Θ_f = 136 ± 3°. The displacement of the ridge node is accommodated by a change in length of the adjacent ridges that form the node. Similar results have been obtained by displacing other ridges on a variety of different FLG samples. A reasonable conclusion drawn from these experiments is that the surface of the C face FLG contains a flexible network of secondary interlocking ridges, rather than a rigid network that is locked in place. The ridge network is similar to wrinkles in a rug, hastily arranged on a hard slippery floor. Much in line with the analogy of the rug, the ability to move secondary ridges in FLG indicates that the interface between the FLG and SiC exhibits weak bonding.

3.4. Force versus lateral displacement of a secondary ridge

In order to estimate the forces required to displace a ridge, a sequence of experiments were performed in which the normal and lateral forces as a function of displacement were measured.
Figure 5. A close-up AFM image of a ridge node found in FLG grown on hydrogen-etched 4H-SiC(0001) at 1550 °C for 10 min. The ridge height is about 10 nm. From figure 1, we infer an FLG thickness of ~11 layers of graphene. In (a), an AFM image of a ridge network before performing a nanomanipulation experiment is shown. In (b), the normal and lateral forces as the tip execute a contact mode scan across a region of the flat FLG substrate. In (d) are shown the normal and lateral forces when the tip executes a contact mode scan across the ridge, as indicated by the white arrow in (a). A distinct, nearly symmetric feature is observed in both the lateral and normal forces. In (c), the AFM image after the contact mode line scan shows no evidence for ridge deformation.

while the tip executed a contact mode scan across a ridge. In many cases, the applied lateral force was insufficient to displace a ridge. An example of such an experiment is given in figure 5. An AFM image of a ridge network with a height of ~10 nm found on a 1550 °C FLG sample is given in figure 5(a). Figure 5(b) plots the normal and lateral forces when the tip executes a contact mode scan across a flat region of the FLG surface. During the lateral displacement of the tip, a signal proportional to the lateral force was measured by digitizing the lateral force signal from the AFM photodiode. From these data (in volts), an estimate for the lateral force (in µN) was obtained, as discussed in section 2.2. The ridges are quasi-rigid features on the surface and can be displaced when the lateral force exceeds a threshold value required to move the ridge. Figure 5(c) shows this particular ridge resisted displacement, even after applying a normal force of ~1 µN perpendicular to the ridge axis. The corresponding lateral force is not sufficient to displace this particular ridge. Under these circumstances, a rather symmetric feature is observed in both the lateral and normal forces when the tip scans across the ridge (see figure 5(d)).

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Figure 6. A close-up AFM image of a secondary ridge node found in FLG grown on hydrogen-etched 4H-SiC(0001) at 1550 °C for 10 min. The ridge height is about 4 nm. From figure 1, we infer an FLG thickness of ~11 layers of graphene. In (a), an intermittent contact AFM image obtained under ambient conditions is shown. A white line with points P₁, P₂, P₃ and P₄ indicates the location of a contact mode scan (from left to right) performed during a nanomanipulation experiment. In (b), an AFM image after performing the nanomanipulation experiment is shown. The closed ridge node evident in (a) has clearly opened in (b). The bottom panels show normal and lateral force data acquired during the nanomanipulation experiment. While the normal force is reasonably constant over the entire contact mode scan, the lateral force shows a significant linear increase between the points P₂ and P₃ before returning to its nominal value. These data suggest that the ridge is displaced roughly as expected from a Hook’s law model and then snaps back to approximately its original position. In (c), an AFM image obtained after a similar contact mode scan is performed in the reverse direction (from left to right) is shown. The image shows the restoration to a closed ridge node.

The small linear change in the normal and lateral forces observed in figures 5(b) and (d) is attributed to the angular bending of the z-piezo used to move the sample when a stationary tip is dragged along the pre-specified line (white arrow in figure 5(a)).

The results of a similar experiment on a different ridge network are summarized in figure 6. In this case, we have evidence that the ridge is displaced laterally, but then snaps back to a location close to its initial position. The secondary ridge in figure 6(a) has a height of ~4 nm and was found on a 1550 °C FLG sample. The white line in this figure indicates the path of the tip in contact mode as it was swept across the ridge. The experiment was repeated a number of times, applying an increasing normal force. In all instances, no evidence for a deformation of the ridge was observed until a normal force of 907±13 nN was reached. In figure 6(a), four relevant points are indicated by the letters Pⱼ, j = 1, 4. The bottom panel shows that the normal force is reasonably constant at 907 nN during the entire 600 nm long contact mode sweep.
During the lateral displacement of the tip, a signal proportional to the lateral force was also measured. The results of this experiment are also plotted in the lower panel of figure 6, where the average lateral force is inferred to have a reasonably constant value of \( \sim 30 \) nN.

Figure 6(b) shows the AFM image of the ridge after the contact mode scan was complete. Clearly, the node structure has changed from a closed to an open triangle-like structure. The lateral force data in figure 6 also exhibit an unmistakable linear increase in the lateral force during a portion of the contact mode sweep. In particular, a significant increase in the lateral force is observed between points P\(_2\) and P\(_3\), indicating that the change in lateral force occurs when the tip first comes into contact with the secondary ridge. The linear increase in lateral force as the tip displacement increases suggests that a Hook’s law model is appropriate to describe the lateral ridge displacement. A lateral spring constant of \( \sim 0.13 \) Nm\(^{-1}\) is estimated from the data. After the point P\(_3\), the lateral force abruptly returns to a constant value consistent with the initial lateral force at the start of the contact mode scan.

The linear increase in lateral force as a function of position suggests that the ridge is displaced by the tip from point P\(_2\) to P\(_3\). Near the point P\(_3\), the elastic restoring force of the ridge exceeds the lateral force applied by the tip and the ridge evidently snaps back to its original position. While snapping back, the displacement is not entirely elastic as indicated by the permanent deformation of the ridge node, as is evident by comparing figures 6(a) and (b).

To complete this particular nanomanipulation study, the tip executed a reverse contact mode scan in the opposite direction. The resulting AFM image after this experiment is presented in figure 6(c) and indicates that the node structure has been returned to its original, closed shape.

4. Discussion

As determined from the AFM studies discussed above, the graphitic layer that forms on the surface of the C face of 4H-SiC contains flat tile-like regions of FLG surrounded by a highly deformed, 1D ridge-like mesh network. The physical characterization and origin of this ridge network have been discussed elsewhere \([19]\). Briefly, after FLG growth at high temperature, anisotropic compressive stress develops upon cooling due to the differential thermal contraction between SiC and the graphitic overlayer. In response to this compressive stress, the FLG film deforms and buckles, forming ridges. At this time, we do not conclusively know whether this buckling occurs due to intrinsic corrugation fluctuations (rippling) in the graphene layers \([26]\) or whether it is seeded by point-like defects in the FLG that form during graphene growth. The driving force for the formation of ridges (or folds) is the minimization of the strain energy in the FLG film. Ridges may likely form during cool down when wrinkles in the FLG coalesce. The ridges tend to align in directions determined by the weakest points in the FLG film, in much the same way that cracks propagate following directions of least resistance. Whether a ridge can be displaced or not depends on local conditions. Two secondary ridges with nominally the same dimensions can exhibit different deformation properties when pushed laterally by a tip.

In order to better understand the atomic character of the ridge structure in FLG, high-resolution TEM studies were also conducted. The sample preparation techniques are summarized in section 2.3. A TEM micrograph of the cross-sectional area of a ridge found in an FLG sample grown at 1600 °C is given in figure 7. The height of the ridge is about 5 nm and from this value we infer that the TEM image is of a secondary ridge in the FLG. Furthermore, from figure 7, the FLG–SiC interface can be rather well identified. The average FLG thickness
Figure 7. A TEM image of a secondary ridge in cross-section found in FLG grown on hydrogen-etched 4H-SiC(0001) at 1600°C for 10 min. The ridge height is about 5 nm. By direct count of the graphene layers, the TEM image indicates ~11 graphene layers, in reasonable agreement with the XPS thickness of ~15 layers inferred from figure 1. The image provides supporting evidence that the ridge forms from a delamination of the entire FLG from a specific region of the SiC substrate. Careful examination of the graphene layers in the ridge provides clear evidence for atomic-scale corrugation that relieves stress during ridge formation.

is found to be 5 nm and, using the $c$-axis plane separation of 0.335 nm, we estimate that 14.9 layers of graphene make up the FLG thin film. A direct count of the graphene layers provides a value close to 11 layers. For the sake of comparison, at this growth temperature, we would estimate that ~15 layers of graphene form from the results given in figure 1. These results are all in reasonable agreement with each other.

An examination of figure 7 provides clear evidence for the delamination of the entire ridge from the substrate and, furthermore, shows that the ridge height is comparable to the ridge width. Good evidence for corrugations in the ridge is seen. These corrugations provide corroborating evidence for similar structures inferred from high-resolution STM scans [20].

5. Conclusions

Hydrogen-etched 4H-SiC(0001) substrates were held at elevated temperatures under high vacuum conditions for 10 min. After cooling to room temperature, studies of the mechanical characteristics of the ridge network that forms on the carbon face of the SiC substrates as a
function of growth temperature were performed. A variety of FLG samples grown at different temperatures were studied and no systematic differences were observed between the different samples investigated. Specifically, an interlocking mesh of ridge-like features was found to surround tile-like regions of atomically flat FLG for every sample studied. The ridge network serves to roughen the surface of FLG in an uncontrolled way. The origin of the ridge-like network is attributed to the 2D compressive stresses that develop upon cooling from the growth temperature. The precise shape of the ridge-like network is driven by intrinsic buckling instabilities of the FLG and is likely influenced by the random location of atomic-scale point-like defects that form during graphene growth.

Both primary and secondary ridges were identified and studied using the AFM force versus z-displacement data and nanomanipulation techniques. The normal force versus z-displacement experiments revealed a ridge that was substantially more compliant than the surrounding flat FLG substrate. Lateral force experiments showed that secondary ridges can be permanently deformed by contact mode scans. Typically, high normal forces (>$0.5\ \mu\text{N}$) are required to displace the secondary ridges, while the primary ridges remain locked for the range of forces that have been investigated. In some instances, evidence for elastic ridge snap-back after displacement has been seen. High-resolution TEM studies provide direct evidence that the ridge is formed by the delamination of the entire FLG layer from the SiC substrate. Taken together, these experiments provide compelling evidence for a weakly interacting FLG–SiC interface.

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