Temperature dependence of molecular beam epitaxy of GaN on SiC (0001)

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Abstract

High quality gallium nitride (GaN) thin films have been grown on 6H-silicon carbide (0001) substrates at varying substrate temperatures using molecular beam epitaxy and characterized at low and high film thicknesses. The epitaxial layers show two regimes in temperature distinguishable by different morphology. For film thicknesses around the critical thickness, low temperature growth is two dimensional while for higher temperatures, growth is in the form of 3-D columnar islands. At a thickness of about 200 nm, the low temperature films show a large density of spiral growths while high temperature films show a 2-dimensional morphology. X-ray peak widths are seen to decrease with increasing substrate temperature. These results have been explained in terms of a model which proposes different misfit dislocation formation mechanisms in the two temperature regimes.

1 INTRODUCTION

Gallium nitride (GaN) is a wide bandgap semiconductor with direct bandgap which makes it attractive both as a short wavelength light emitter and a high temperature electronic material. There are no easily available, stable, lattice-matched substrates for this material for device development and so heteroepitaxy has been performed on sapphire (mismatch of about 15 %) and silicon carbide (SiC, mismatch of about 3 %) [1]. It is expected that SiC with a smaller mismatch would lead to heteroepitaxial GaN of higher quality than sapphire, but several factors other than mismatch are believed to be reasons for GaN films on SiC not being significantly better [2].

In this work, we have grown GaN on Si-polar SiC(0001) using molecular beam epitaxy (MBE). Substrate temperature is varied for different films and the structural differences arising from this are reflected in the surface morphology and high resolution x-ray diffraction (HRXRD) peak widths.

2 EXPERIMENT

Si-face 6H-SiC(0001) surfaces for our experiments are prepared by removal of polish damage from the wafers using hydrogen etching [3]. This technique is known to produce large, flat areas on the wafer suitable for performing STM experiments on. After introducing the substrates into ultra high vacuum (UHV, pressure less than 10^{-10} Torr), the samples are outgassed at about 800°C for 30 minutes. Prior to desorption of the native oxide layer, Si is deposited on the substrate using an electron beam source. This replenishes any Si that may be lost by the surface during the oxide removal step. Oxide desorption is done by annealing the substrate at about 1000°C till a 3×1 reflection high energy electron diffraction (RHEED) pattern (indicative of a $\sqrt{3}\times\sqrt{3}$-R30° surface reconstruction) is obtained.

GaN films of up to 200 nm thickness are grown by MBE on this substrate using a Ga effusion cell and an RF-plasma nitrogen source. The growth is a single-step process. The substrate temperature is varied for different growth runs and is monitored using a pyrometer and a thermocouple in contact with the back of the sample mounting stage. Growth is performed under highly Ga-rich conditions as indicated by the streaky, dim RHEED pattern during growth [4].

UHV GaN film characterization is performed by in-situ RHEED and scanning tunneling microscopy (STM). Outside UHV, we perform atomic force microscopy (AFM) and high resolution x-ray diffraction (HRX11RD) measurements on the samples.

3 RESULTS AND DISCUSSION

After the growth process, the samples are allowed to cool. When the sample temperature is between 200° C and 300° C, all the samples show a pseudo-1×1 RHEED pattern as described previously [5,6]. This RHEED pattern is shown in Fig. 1 (a). Excess Ga on the surface is desorbed by holding the sample at 650° C for a few minutes. The RHEED pattern after heating is 2×1, and occasionally 2×2, as shown in Figs. 1 (c) and (d). Deposition of Ga and subsequent flashing at 650° C lead to 4×6 and 5×5 RHEED patterns. A photograph of the 4×6 is shown in Fig. 1 (b). The Ga content of the surface decreases from Fig. 12 (a) to (d). All these RHEED patterns are seen specifically on Ga-polar GaN [5]. Moreover, the surface is resistant to etching by NaOH, which is further indication that GaN grown on Si-polar SiC(0001) is Ga-polar [7]. This conclusion is in agreement with previous experimental results [8] and theoretical predictions [9].

Figure 2 (a) shows the STM image of a thin (about 4 nm thick) film of GaN grown at a substrate temperature of 550° C. The surface is seen to be almost flat and made up of terraces that show sub-angstrom rippling. Rippling on the terraces arises from the presence of misfit dislocations at the GaN-SiC interface [10]. Since the misfit strain is compressive, misfit dislocations will be created by absent rows of atoms at the interface and lead to dips in the surface above them. This causes the appearance of dark filamentous features which constitute the rippling on the terraces. Also in this figure, labeled S, are a pair of screw dislocations emerging at a step edge. These will eventually give rise to spiral growth fronts in the sample.

Shown in Fig. 2 (b) is the STM image of a GaN film with thickness similar to that of Fig. 2 (a), grown in this case at 650° C. In contrast with Fig. 2 (a) where the image greyscale is 6.5 Å, the greyscale of Fig. 23 (b) is much larger, 27.3 Å. Growth appears to have taken place in the form of 3-dimensional columnar islands as is shown by the deep crevices that are seen on the surface. In this image, the islands are seen to be in the process of coalescing.

The temperature dependence apparent in Fig. 2 for GaN MBE growth is similar to that seen in Si-Ge/Si heteroepitaxy [11]. For the Si-Ge/Si system, as Ge composition in the overlayer increases or as growth temperature increases, the growth tends to shift from planar to three dimensional growth. At lower temperature, the overlayer prefers to reduce its surface energy by going into a high strain configuration and at higher temperature, the film tends to decrease its strain energy at the expense of surface energy.

Figure 3 shows high resolution STM images of thin films (thickness ≈ 4 nm) of GaN grown on SiC at 550° C. Several types of defects created by strain relief are seen in these images. All these surfaces show the pseudo-1×1 reconstruction. Accounting for the errors arising from variation in tip length (about 10%), the corrugation maxima are 3.4±0.25 Å apart. This value agrees reasonably well with prior observations [6]. In Fig. 3 (a), two dark linear features have been labeled MD. Two long misfit dislocations at the GaN-SiC interface give rise to these lines as described above. Also labeled in this image is E, an edge dislocation emerging from the plane of the film. A magnified view of E is shown in Fig. 3 (b). In this image, a Burgers circuit has been drawn around the dislocation to show its lack of closure and hence determine the Burgers vector $\vec{b}$ for the dislocation. The vector \vec *b* is also displayed in the figure. It is seen to lie in the plane of the film and has a magnitude equal to one lattice spacing.

Figure 3 (c) shows a pair of screw dislocations S emerging at the surface of the thin film. These may arise from a misfit dislocation at the heterointerface. At the surface they give rise to a step that terminates at the dislocations. As growth proceeds, the dislocation pair may evolve into a pair of spiral growth fronts. On the region around the dislocation pair, several other kind of defects are seen. The triangular depressions in the surface are prismatic dislocations. These occur when a sub-surface prism of material within the triangular boundary is displaced relative to the material around it in a direction perpendicular to the plane of the film. The label P on this defect actually points to a corner of the triangular boundary. This corner is magnified in Fig. 34 (d). In the magnified image, the pseudo-1×1 reconstruction is clearly visible. It is also seen that the edge of the triangle is not an abrupt step, but a gradual dip on the surface.

After removing the samples from the UHV chamber, AFM and HRXRD measurements are made. In Fig. 4 , we show AFM images of 200 nm thick samples grown at 550° C in (a) and at 650\circ C in (b). Figure 4 (a) shows a large number of bright spots, one of which is magnified in the inset. The inset shows that the bright spots are actually spiral growth mounds. Growth at 550° C leads to a large concentration of spiral growths as compared to growth at 650° C. Spiral growths are known to arise from screw dislocations emerging at the film surface. If the smaller number of spiral growth fronts seen in the higher temperature sample is due to a smaller number of screw dislocations in the bulk, then we expect this film to show better structural quality than the low temperature film. At 650° C, the surface is seen to have a two-dimensional appearance. This is also seen in the factor of 5 difference between the greyscales of (a) and (b). AFM images of films grown at 600° C show an intermediate concentration of surface spiral growth fronts.

HRXRD results for films grown at different temperatures are shown in Fig. 5. The first conclusion that can be drawn from this data is that, structurally, these GaN films are of very high quality with rocking curve full width at half maximum (FWHM) between 1 and 2 arc-secs. Both the double crystal rocking curves and triple crystal x-ray diffractograms showed Pendellosüng fringes for films 200 nm in thickness, further confirming the high quality of the material. The double-crystal measurements show a trend of decreasing FWHM for the GaN(0002) peak with increasing growth temperature as seen in Fig. 5 . Assuming that the dislocation systems and their distributions are similar in films grown at any temperature, theories of x-ray diffraction [12,13] predict that dislocation density is approximately proportional to the square of the x-ray linewidth. Estimates based on these approximate theories give the density of dislocations in the film at 550 °C to be about two times greater than the density for films grown at 650 °C.

This difference in dislocation density can be partly be explained in terms of the film morphology during early stages of growth. For the low temperature, 2-D mode of growth, when the film attains critical thickness and misfit dislocations need to be created to relieve strain, dislocations can start on the terraces, step edges or the edges of sample, because dislocations can only end at a surface or in a loop [14]. Thus for every segment of misfit dislocation formed, the formation of two threading dislocations is necessitated. In the case of the high temperature growth, when the film reaches critical thickness it is made up of columnar islands, and the edges of these islands are another set of possible surfaces for dislocation termination [14]. Assuming that the number of misfit dislocation segments formed is the same as at the lower temperature, several of the segments can terminate at the island edges without the formation of threading dislocations prior to island coalescence. When islands begin to coalesce, many of the dislocations formed at the edges of islands will be misaligned and will produce two threading dislocations per pair of misaligned misfit dislocations. There will still be some dislocations terminating at neighboring island edges which will be aligned and during coalescence, will lead to longer segments of misfit dislocations. As islanding becomes more severe, a larger number of dislocations can terminate at island edges and we expect the number of threading dislocations to become smaller. When the size of islands becomes smaller than the length of misfit dislocation segments, we expect the threading dislocation density to rise again because of an increasing number of misaligned misfit dislocations during island coalescence.

We believe the increase in FWHM at 700 °C seen in Fig. 5 is not a consequence of the severe islanding phenomenon mentioned above, but may be due to other kinds of bulk defects formed in the Ga poor growth regime caused by the high substrate temperature. At the end of growth and cooling, for the samples grown at 700 °C, the RHEED pattern seen is the relatively low Ga content 5×5 which is indicative of Ga poor growth. These samples also appeared yellower than any of the others leading us to the conclusion that they have a higher defect density.

The x-ray FWHM measured for all these films is close to the peakwidth expected from the diffraction pattern caused by the finite thickness of the films, therefore further x-ray studies are in progress to understand the defect structure of thicker films. These studies (to be published elsewhere) show similar trends for the FWHM of x-ray curves with growth temperature variation, but present further details of the dislocation defect structure for these films.

4 CONCLUSIONS

We have grown high quality GaN on hydrogen-etched 6H-SiC (0001) substrates by MBE. The substrate temperature has been varied during growth and the effect of different growth temperatures on morphology and structural quality has been studied. Over the range of temperatures at which the experiments were performed (500–700 °C), the GaN films are observed to be Ga-polar as determined by surface reconstruction and etching behavior in NaOH.

At low GaN film thickness, films grown at lower temperature show 2-D growth while those grown at higher temperature show islanded growth, analogous to Si-Ge on Si heteroepitaxial systems. For films that are about 200 nm thick, both AFM images and x-ray diffraction measurements show a higher dislocation density for low temperature films. A possible explanation for these differences in the thick films has been suggested based on the low thickness morphology. For 2-D growth at low temperatures, termination of misfit dislocations can happen only at the film surface or step edges accompanied by the formation of threading dislocations. In 3-D growth, the edges of islands present more free surfaces for misfit dislocation termination. If dislocations which terminate at neighboring island edges are misaligned with each other, they produce threading dislocations during island coalescence but if they are aligned, they form a longer misfit dislocation segment, and so a smaller total number of threading dislocations.

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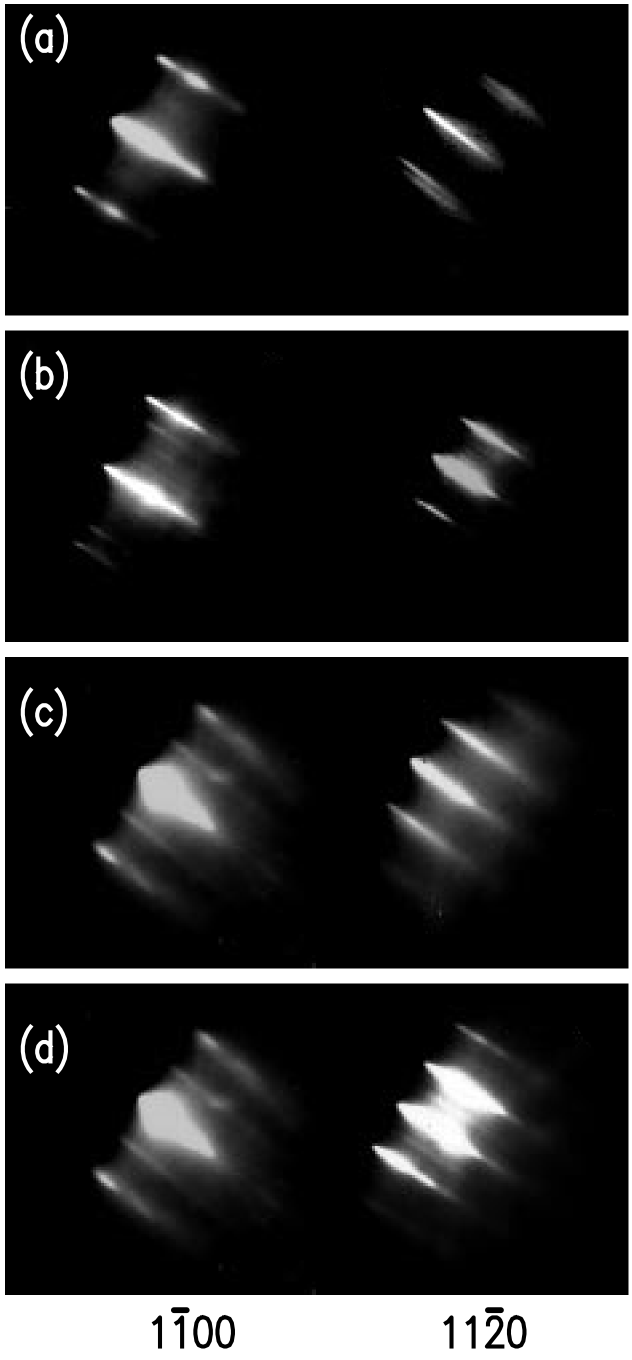

Figure 1 RHEED patterns in the [$1\bar{1}00$] and [$11\bar{2}0$] directions for GaN on 6H-SiC (0001) shown in order of decreasing surface Ga content (a) pseudo 1×1, (b) 4×6, (c) 2×1 and (d) 2×2.

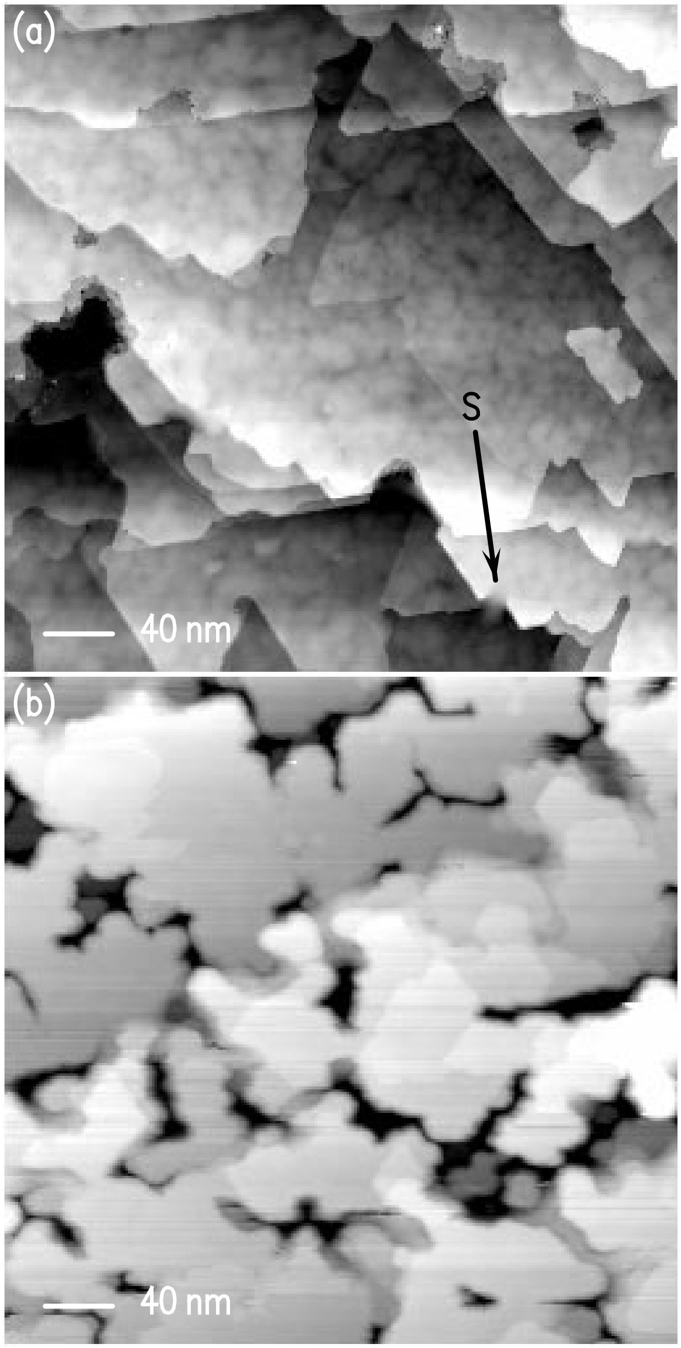

Figure 2 (a) STM image of a GaN film about 4 nm thick grown at 550° C. S labels a pair of screw dislocations emerging at the film surface.(b) Similar thickness GaN film grown at 650° C.

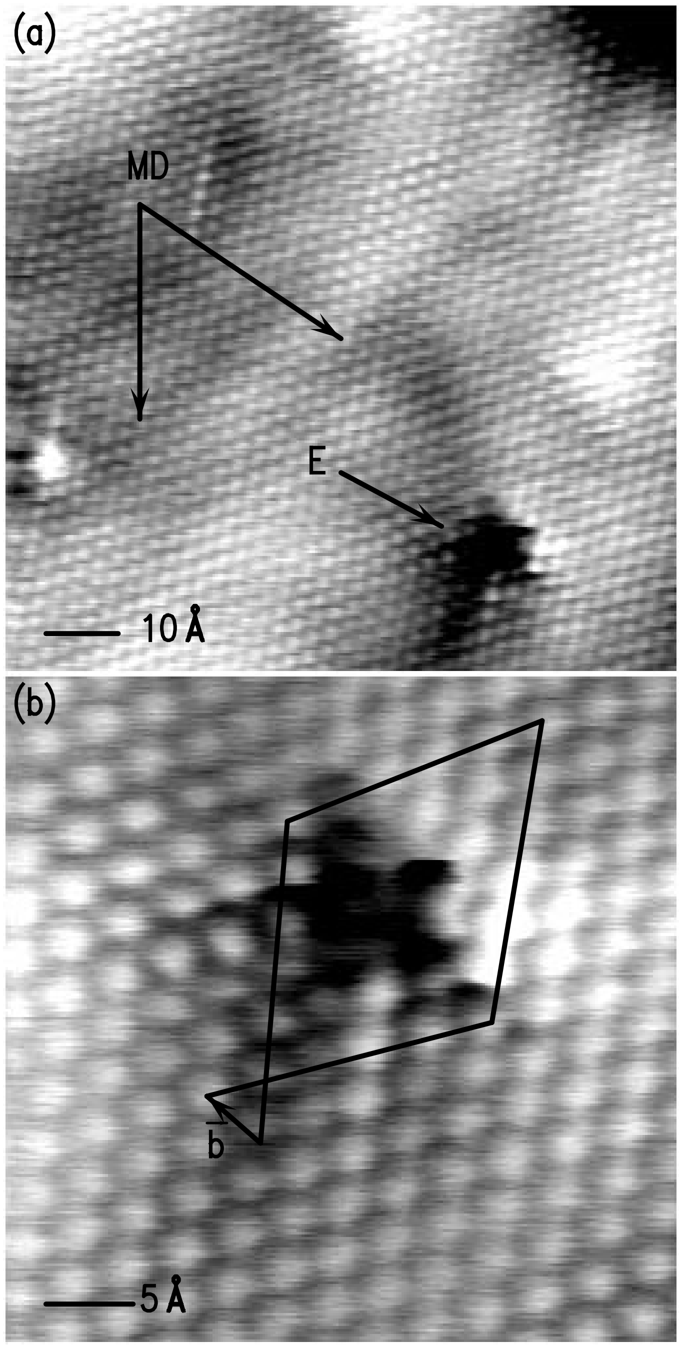

Figure 3 High resolution STM images of a thin GaN film grown at 550 ° C. (a) Underlying misfit dislocations give rise to the dark features labelled MD. E is an edge dislocation, shown magnified in (b).

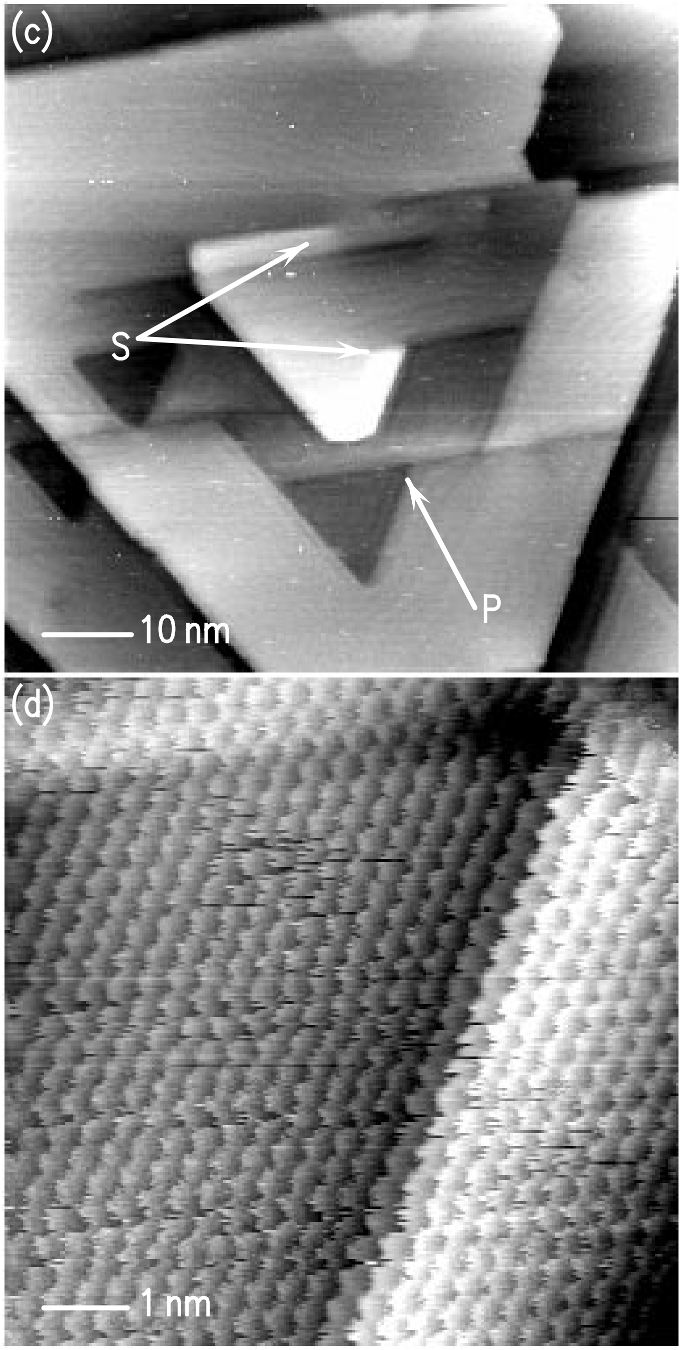

Figure 3 (c) Pair of screw dislocations S emerging at the film surface. P is a prismatic dislocation, whose corner is magnified in (d). Greyscales of the images are 0.5, 0.6, 2.5 and 0.9 Å respectively.

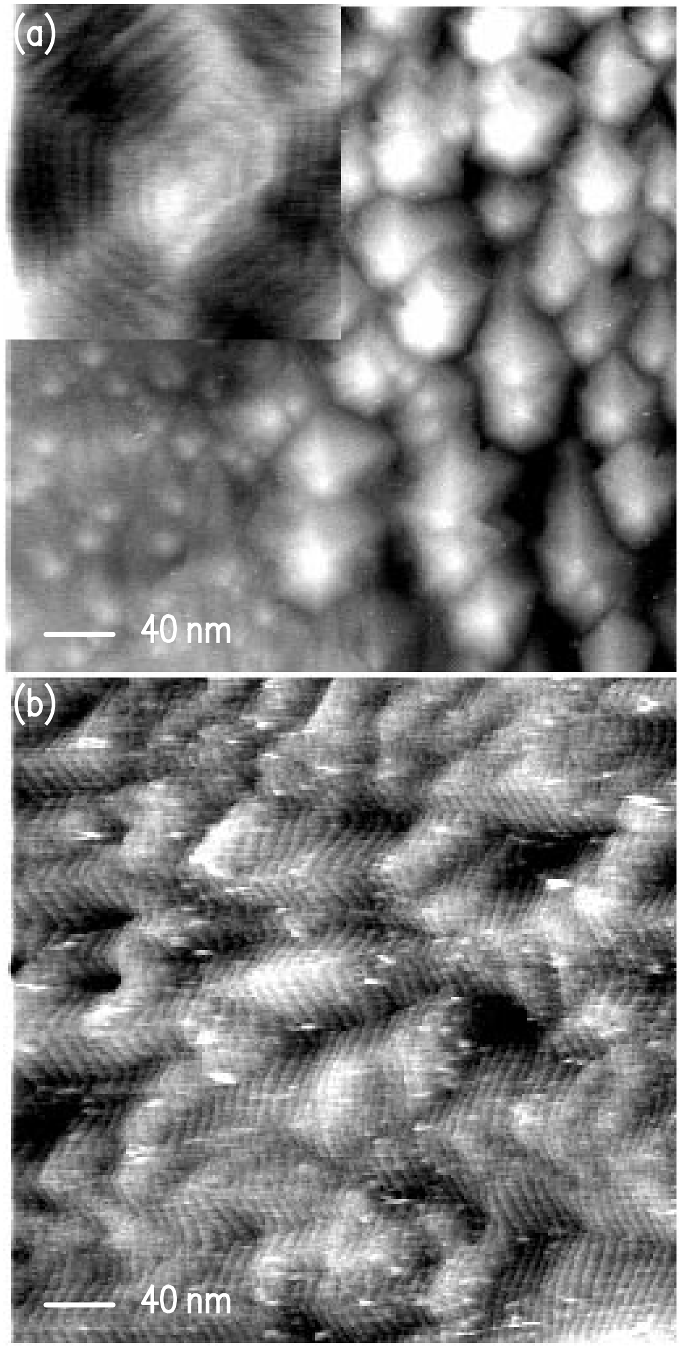

Figure 4 (a) AFM image of 200 nm thick film of GaN grown at 550° C. The 0.5µm×0.5µm inset shows a close-up of a spiral growth mound. Greyscales of the large image and the inset are 83 Å and 20 Å respectively. (b) AFM image of a 200nm thick GaN film grown at 650° C. Greyscale for this image is 15.9 Å.

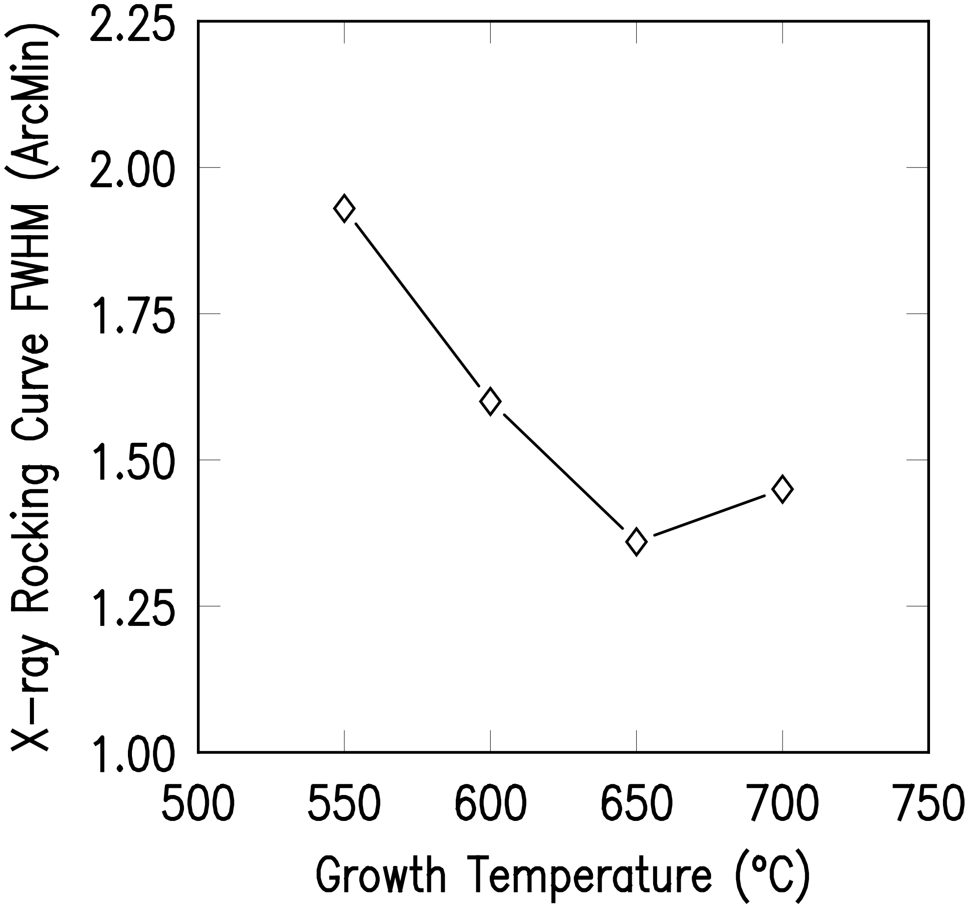

Figure 5 HRXRD double-crystal linewidths of the α-GaN (0002) peak as a function of temperature.