Large scale surface structure formed during GaAs (001) homoepitaxy

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Atomic force microscopy studies have been performed on GaAs (001) homoepitaxy films grown by molecular beam epitaxy. Multilayered features are seen to evolve when the growth conditions favor island nucleation. As the epilayer thickness is increased these features grow in all dimensions but the angle of inclination remains approximately constant at 1°. The mounding does not occur on surfaces grown in step flow. We propose that the multilayered features are an unstable growth mode which relies on island nucleation and the presence of a step edge barrier.

GaAs surfaces grown using molecular beam epitaxy (MBE) have produced devices of the highest quality. Because of its practical importance and because the model for its growth poses an interesting intellectual problem, these MBE grown surfaces have been widely examined by a variety of techniques. Reflection high-energy electron diffraction (RHEED) has proven to a particularly powerful probe giving real time information on growth mode transitions, surface diffusion lengths, and surface reconstruction as the film develops. More recently scanning tunneling microscopy (STM) has been used to obtain real space information at various points during the evolution of the surface. Both of these techniques are capable of, and in fact specialize in, giving atomic scale details about the surface but they can only hint at larger features.[1-4] In this letter we will focus on large scale features using an atomic force microscope (AFM) to obtain topographic information on a scale larger than that of either a typical STM which has a scan range of 500 nm or RHEED which has a coherence length of approximately 100 nm. We find that during growth under certain conditions multilayered mounds form. These features have appeared in the literature having been observed by laser light scattering (LLS),[5,6] reflection electron microscopy,[7] and AFM.[6,8] In this letter we explore the early development giving a physical argument for their evolution and the growth conditions needed to prevent their formation.

Our samples were grown on GaAs (001) substrates with a variety of miscuts ranging between 0.1° (nominally singular) and 2°. The typical As/Ga pressure ratio was 10. The growth rate was approximately 0.18 μm/h. The substrate temperature during growth was varied between 555 and 620 °C as measured with a pyrometer. Before loading the samples into the vacuum chamber, the substrates were cleaned using a standard HCl etch. Once in the growth chamber the oxide layer was desorbed under an As_4 flux prior to growth. A more detailed account of the experimental apparatus and growth techniques has been published previously.[9]

We used a force microscope with commercial cantilevers which have a pyramidal stylus and a 40 nm tip radius. The samples were imaged in air and therefore have a thin oxide layer. One surprising result is that single bilayer steps (0.28 nm) can be easily seen on smooth terraces. The lateral resolution may be reduced due to the oxide but none of the images shown in this letter challenge this limit. The surfaces

remained unaltered over many scans indicating that we were not perturbing the surface as we traced it with the probe.

The first series of images, Fig. 1 shows how the surface of a singular substrate evolves as increasing thicknesses of GaAs films are deposited on it. For this experimental se-
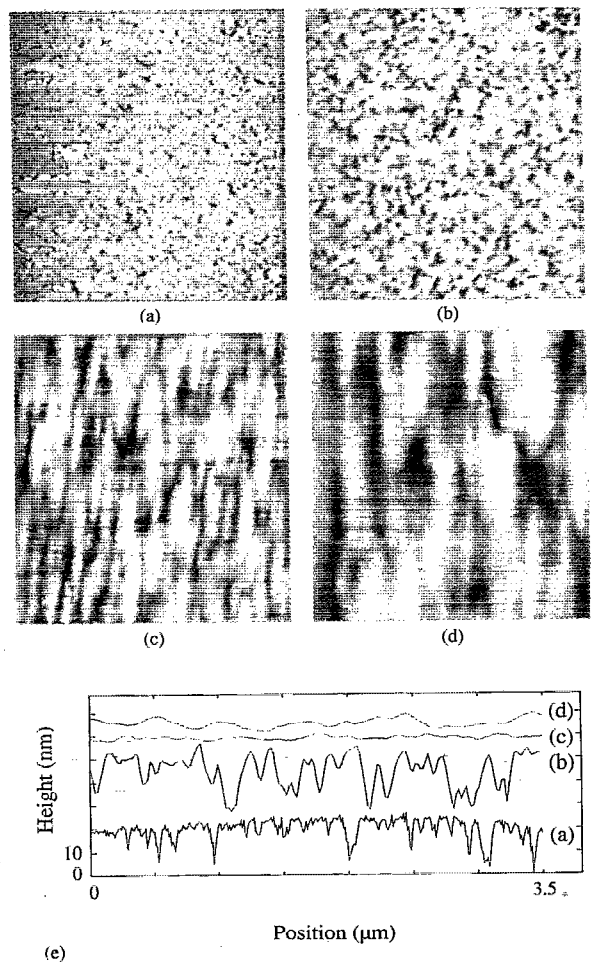

(a) (b)

(c) (d)

(e)

FIG. 1. 3.5 μm×3.5 μm AFM images of homoepitaxy GaAs for various film thicknesses. The rms roughness over this scan is given for each image. (a) Substrate after oxide desorption: rms=3.9 nm. (b) 90 bilayers: rms=6.8 nm. (c) 270 bilayers: rms=0.7 nm. (d) 540 bilayers: rms=1.6 nm. (e) Cross sections of images (a)–(d) with an arbitrary offset between each. The scale on the y axis is 10 nm per division.

quence the substrate temperature is fixed at 555 °C and the growth is quenched after depositing 25, 75, and 150 nm of material. The initial substrate (post-oxide desorption) is relatively rough. The surface has pits with lateral size 20–200 nm and depths 5–25 nm. After 25 nm have been deposited [Fig. 1(b)], the small scale roughness has decreased but the large pits remain.

Figure 1(c) shows the surface after 75 nm have been deposited. The surface is now considerably smoother than either of the two previous images as is demonstrated by the line scan [Fig. 1(e)]. The surface is no longer pitted and has developed elongated multilayered features with their major axis along the [$\bar{1}10$] direction. It is on these gross features that we would like to focus our discussion.

The mounds in Fig. 1(c) have a typical size of 1.0×0.25 μm and are eight or nine bilayers. The average angle of inclination between the sides and the average plane is approximately 1° along the [110] direction. As more layers are deposited, the features become larger but the angle of inclination remains constant. Figure 1(d) shows the surface after deposition of 150 nm. Mounds are observed with typical dimensions 1.5 μm$\times$0.37 μm$\times$35 Å.

Data consistent with these features have been reported by several groups. Briones et al.[5] observed different scattered light intensities along the [$\bar{1}10$] and [110] directions which they attributed to a "highly anisotropic surface ripple." More recent LLS experiments have been carried out in situ[6,10] and have shown that the scattered light along the [110] direction increases with epilayer thickness implying that a surface feature is growing. Smith et al.[6] confirm this with AFM images of GaAs with epilayer thickness from 0.2 to 2 μm. Our images (Fig. 1) show this for the earlier development of the mounds. Scanning reflection electron microscopy[7] has also shown that an anisotropic undulation develops as the film thickness is increased above some nominal thickness (>20 nm for their growth conditions). This can be seen clearly by the change in morphology between Fig. 1(b) and Fig. 1(c). The first 20 or so bilayers are strongly influenced by the initial highly pitted substrate and it is only after the pits are smoothed that the mounds appear.

This type of mounding represents an unstable growth mode which we believe occurs when the competition between step-flow growth and nucleation favors the nucleation of new islands. Island nucleation occurs when the terrace width, L, is larger than the typical distance an adatom travels before nucleating an island (σ).[11] For two-dimensional islands to turn into multilayered mounds more adatoms must land on top of them than hop off. Thus the tendency to mound requires that the material exhibit a step edge barrier (Schwoebel barrier[12] or diffusion bias[13]) which impedes the motion of an adatom off of a terrace. Such a barrier has been studied with metal films and although it has not been directly observed for GaAs it is reasonable to expect a similar effect. The elongated shape of the mounds is a reflection of anisotropic diffusion or sticking probabilities[2] but anisotropy is not a necessary component for the surface instability.

On a nominally singular surface the terraces are large. Thus for typical growth temperatures the terrace width is larger than σ and the adatom has a high likelihood of nucle-
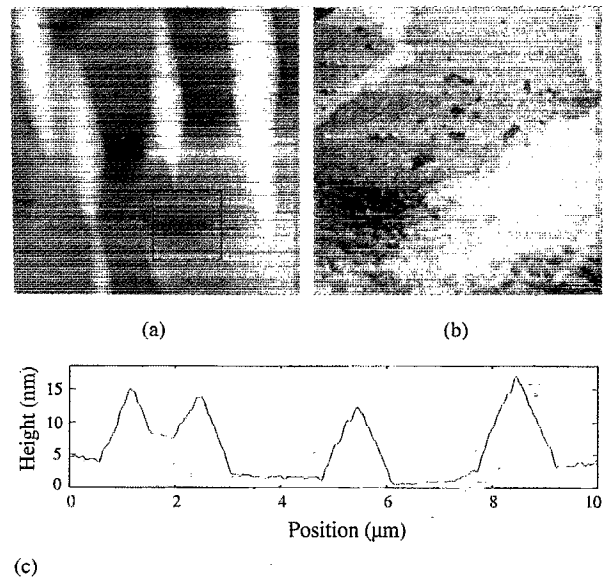

(a)　　　　　　(b)

(c)

FIG. 2. (a) 10 μm$\times$10 μm AFM scan of a 200 nm homoepitaxy GaAs surface after being annealed 30 min at 555 °C. The rms roughness over this area is 3.5 nm. The mounds are 10–15 nm high at their center points and have an aspect ratio of ~5 with a typical size of 6.6 μm$\times$1.3 μm. They are elongated along the [$\bar{1}10$] direction. (b) 2.3 μm$\times$2.3 μm scan of the inset indicated in (a). The rms roughness over this area is 0.4 nm. The upper-half of the image is primarily one very large terrace. The cascading terraces on the lower-half of the image have an average width of 120 nm which corresponds to a slope of 0.1° (the nominal miscut). (c) A cross-sectional cut along the [110] direction. The slopes of the sides are ~1° all along the length of the mounds.

ating an island rather than reaching a step edge. Once an island is large enough to have a significant Schwoebel barrier, adatoms which land on its terrace tend to be reflected away from the edges and hence become trapped on the island, contributing to growth of the next layer. The mounds continue to grow by nucleating new islands on the upper terraces. The sides of the mounds have terrace widths less than σ and therefore grow by step flow, the bottoms merge with other mounds as they grow closer together. This growth scenario has been numerically simulated and found to reproduce the qualitative results of the experiment. Monte Carlo results will be published elsewhere.[11,14]

This mounding can be seen in a spectacular way on a 200-nm-thick partially annealed surface. Here the smaller mounds have been absorbed into the larger features leaving a very flat surface with 10-nm-high ridges spaced 1–3 μm apart. Although mounds of this magnitude have not been documented in the STM literature, this type of surface does not contradict STM observations. This image is largely composed of broad terraces which on a STM scale would seem endless. An enlargement of a section between the mounds [Fig. 2(b)] shows a region 2 μm by 2 μm with only four bilayers present. Thus a representative 0.2 μm image for this sample might either show large, flat terraces or it might show regular steps with terrace widths of ~15 nm.

If the temperature of the substrate during growth is increased or if the terrace width is decreased an adatom becomes more likely to incorporate into a step edge rather than to nucleate an island. Thus, on miscut samples with substrate
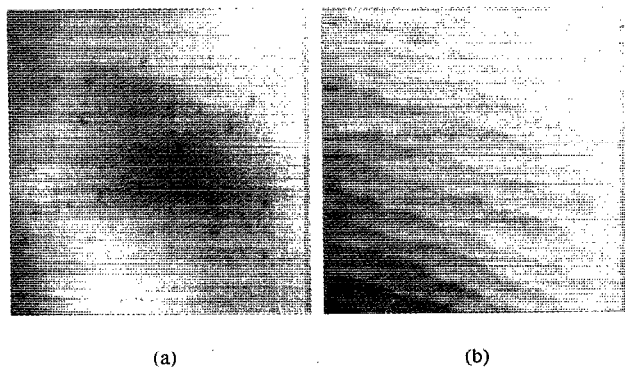
(a)   (b)

FIG. 3. AFM images of homoepitaxy GaAs after deposition of 200 nm on a 0.3° miscut substrate. The substrate temperature during deposition was 620 °C which resulted in step-flow growth. No mounds were found on this sample. (a) 11.5 μm×11.5 μm scan exhibiting no large scale features. (b) 0.5 μm×0.5 μm scan which shows the surface to be in step flow. The terrace widths are correct for the nominal miscut of the surface.

temperatures sufficiently high for the growth to proceed by uniform step flow, we expect no mounding. This is corroborated by the observation that for samples grown at high substrate temperatures LLS intensity is reduced and further the anisotropy disappears indicating the absence of mounds.[5]

An example of this type of growth mode can be seen in Fig. 3 for a nominally 0.3° miscut sample grown with a substrate temperature of 620 °C. The average terrace widths are 44 nm which correspond well to the nominal miscut. The depressions in image (a) are due to step pinning which we believe is caused by impurities during high temperature deposition and will be the subject of a future publication.

As a final test of the model we examined a film deposited on a sample with a thermal gradient. In the hottest region RHEED indicated step-flow growth whereas near the cooler edge island nucleation is expected. Image 4(a) was taken at the center of the substrate (the hottest region) and shows a variation of a few bilayers over a 10 μm×10 μm area. Image 4(b) on the other hand was taken several millimeters away and shows mounding.

We have shown that when growth conditions favor nucleation mounds develop and continue to grow as the epilayer thickness increases. To avoid the formation of mounds the growth must progress in step-flow mode. Additionally, because mounds are not an equilibrium state, they can be annealed away although because they are large, this may take longer than was previously assumed.[6]

We would like to thank C. Gordon, D. Scott, and A. Hunt for assisting with data analysis. C.O. would like to
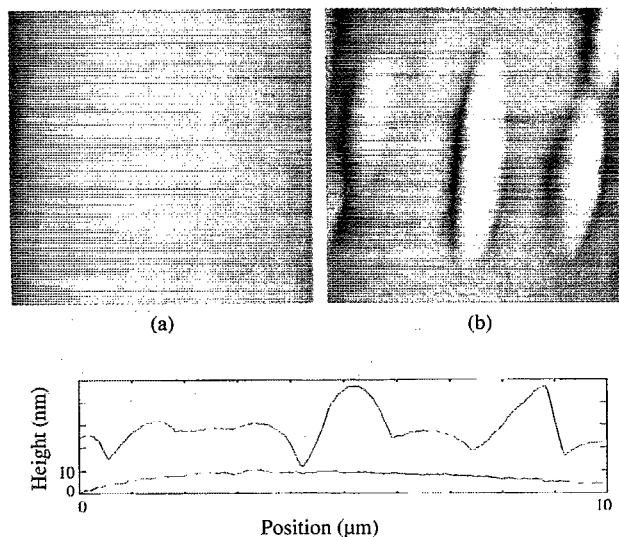

(a)   (b)

Position (μm)

FIG. 4. 10 μm×10 μm AFM scans of a GaAs homoepitaxy on a 2° A miscut sample. (a) was grown in step flow with a substrate temperature of 600 °C whereas (b) was grown cooler due to a thermal gradient on the sample holder. Mounds are observed here.

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