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ATOMIC STEP ORGANIZATION IN HOMOEPITAXIAL GROWTH ON GaAs(111)B SUBSTRATES

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Abstract

When homoepitaxial growth is performed on exactly oriented (singular) $(\bar{1}\,\bar{1}\,\bar{1})$ GaAs substrates, while maintaining the $\sqrt{19}$ x $\sqrt{19}$ surface reconstruction, the originally flat surface spontaneously evolves vicinal $(\bar{1}\,\bar{1}\,\bar{1})$ facets that are tilted approximately 2.5° toward the $<2\,\bar{1}\,\bar{1}>$ azimuthal directions. These facets form pyramid-like structures where the distance between adjacent peaks can be varied from as little as 1 μm to tens of μm. When these surfaces are observed with atomic force microscopy (AFM), we find that they are extremely smooth with the observed tilt resulting from atomic steps which are spaced at approximately 7.5 nm. We have also studied growth on vicinal GaAs$(\bar{1}\,\bar{1}\,\bar{1})$ substrates. Our results are interpreted as indicating that the 2.5° vicinal $(\bar{1}\,\bar{1}\,\bar{1})$ surface has a minimum free energy for the $\sqrt{19}$ x $\sqrt{19}$ reconstruction (i.e., that 10 nm spacing of $<011>$ steps is thermodynamically preferred). Exactly oriented $(\bar{1}\,\bar{1}\,\bar{1})$ facets are only observed when their facet width is less than a couple of micrometers implying a minimum nucleation size. This is a surprising result since conventional wisdom argues the surfaces with low Miller indexes are preferred. A possible explanation is an anisotropy in the surface in the two degenerate phases of $\sqrt{19}$ x $\sqrt{19}$ reconstruction which are rotated $\pm23°$ from the unreconstructed surface.

Key Words: $(\bar{1}\,\bar{1}\,\bar{1})$ GaAs substrates, atomic force microscopy, vicinal GaAs$(\bar{1}\,\bar{1}\,\bar{1})$ substrates, $\sqrt{19}$ x $\sqrt{19}$ reconstruction, surface morphology, strained films, facets, molecular beam epitaxy, step bunching, 2x2 surface reconstruction.

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Introduction

The evolution of surface morphology during crystal growth is an important area of study both for technological applications and for fundamental studies of surface physics. Many applications of epitaxial growth require nearly atomically smooth surfaces although there is also interest in taking advantage of the way some growing crystal surfaces facet to form quantum wires and quantum dots. During epitaxial growth, roughness and/or step bunching can occur for either kinetic or equilibrium reasons; it is appropriate to attempt to understand which dominates. In this paper, we present a detailed study of homoepitaxial growth on the GaAs$(\bar{1}\,\bar{1}\,\bar{1})$ (which is sometime designated as the GaAs(111)B surface in the literature) surface on which spontaneous step bunching is observed. Our experiments indicate that the equilibrium crystal shape is actually tilted some 2.5° away from the $(\bar{1}\,\bar{1}\,\bar{1})$ axis. The atomic step organization which causes this tilt may result from an anisotropic surface stress due to the $\sqrt{19}$ x $\sqrt{19}$ reconstruction. Growth on the $(\bar{1}\,\bar{1}\,\bar{1})$ GaAs surface has attracted attention recently because of the potential applications of the piezoelectric effect in strained films (Smith, 1986; Mailoit and Smith, 1987) and low threshold laser diode applications (Hayakawa et al., 1987) for III-V films grown in this orientation. Prior work (Yang and Schowalter, 1992) has demonstrated that atomically smooth homoepitaxial growth can be achieved on well-oriented GaAs$(\bar{1}\,\bar{1}\,\bar{1})$ substrates by growing in the high-temperature 1×1 reconstruction regime. However, the substrate temperatures required for growth in this regime preclude controlled growth of InGaAs alloys because of In re-evaporation. Growth in the lower temperature $\sqrt{19}$ x $\sqrt{19}$ surface reconstruction regime has proved attractive for this reason. Unfortunately, when homoepitaxial growth is performed on exactly oriented (singular) $(\bar{1}\,\bar{1}\,\bar{1})$ GaAs substrates, while maintaining the $\sqrt{19}$ x $\sqrt{19}$ surface reconstruction, the originally flat surface spontaneously evolves vicinal $(\bar{1}\,\bar{1}\,\bar{1})$ facets that are tilted approximately 2.5° toward the $<2\,\bar{1}\,\bar{1}>$ azimuthal directions. These facets are extremely smooth
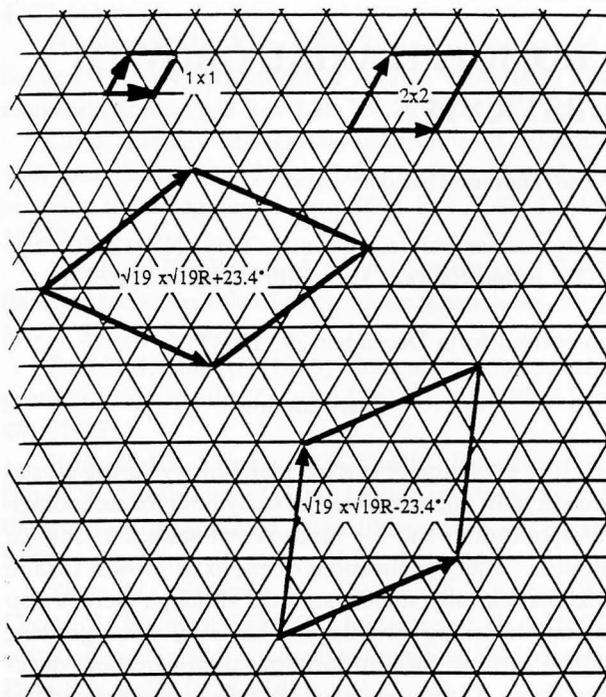
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Figure 1. A schematic of the two-dimensional lattice structure of the GaAs ($\overline{1}\ \overline{1}\ \overline{1}$) surface showing the translation vectors for the 1x1, 2x2, and $\sqrt{19} \times \sqrt{19}$ reconstructions.

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even though they are not aligned with the ($\overline{1}\ \overline{1}\ \overline{1}$) planes indicating that some mechanism for atomic step organization is occurring. For these reasons, we have studied this phenomena in more detail as described below.

Growth

All film growth was done in a Fisons VG90 III-V molecular beam epitaxy (MBE) system (VG Semicon, U.K.) which has a background pressure that is better than 10^{-10} mbar. The surface reconstruction phase was monitored with reflection high energy electron diffraction (RHEED). The GaAs($\overline{1}\ \overline{1}\ \overline{1}$) surface can either exhibit a 2x2, $\sqrt{19} \times \sqrt{19}$, or a 1×1 surface reconstruction depending on the surface As coverage which is determined by the As flux, the Ga flux, and the substrate temperature during MBE growth. The As coverage of the $\sqrt{19} \times \sqrt{19}$ surface is lower than that of the 2x2 surface but higher than that of the 1×1 surface. Details of the surface reconstruction phase diagram have been published previously (Yang and Schowalter, 1992). The $\sqrt{19} \times \sqrt{19}$ reconstruction has two degenerate phases which have unit translation vectors that are rotated by +23° and -23° from the unreconstructed lattice, respectively, as shown in Figure 1. We have always found that these two phases coexist and have approximately the same area as indicated by the RHEED. The

step bunching described in this paper is also always observed for GaAs samples grown in the $\sqrt{19} \times \sqrt{19}$ reconstruction regime. It should be noted that films grown in the 2x2 or the 1×1 regime do not exhibit this spontaneous formation of vicinal facets even when grown on singular GaAs($\overline{1}\ \overline{1}\ \overline{1}$) surfaces.

Our growth experiments were performed with various miscuts of GaAs($\overline{1}\ \overline{1}\ \overline{1}$) substrates. The direction and degree of the misorientation were specified to the substrate manufacturer and were typically checked with Rutherford backscattering/ion-channeling (RBS) measurements. The substrates were typically only within ±0.3° of the nominal miscut specified. The angles reported in this paper should be taken to be of this accuracy. Throughout this paper, we will refer to well-oriented [the surface normal is within ±0.3° of the ($\overline{1}\ \overline{1}\ \overline{1}$) axis] surfaces as singular surfaces to follow the terminology of several theoretical papers on this topic and to emphasize the special character of an aligned substrate.

After growth, the surface morphology of the films has been characterized with optical and electron microscopy. However, most of the quantitative results presented in this paper were taken with an atomic force microscope (AFM). While this AFM is operated in air, it is possible to obtain atomic step resolution (Thundat *et al.*, 1993) with proper control of the room humidity. Care was taken to protect the GaAs surfaces from contamination. However, a gradual degradation of the resolution that could be obtained with the AFM was observed over a period of several months.

Surface Structure

We always observe that growth of GaAs on well-oriented (singular) GaAs($\overline{1}\ \overline{1}\ \overline{1}$) substrates leads to the formation of three-sided pyramids (Yang, 1993; Yang *et al.*, 1993). The main geometric features of the faceted surface morphology can be characterized by two parameters, the tilt angle θ of the facets with respect to the ($\overline{1}\ \overline{1}\ \overline{1}$) crystallographic plane and the distances between the adjacent pyramids d. Typically, θ is found to be somewhat greater than 2° while d ranges from 1 to 30 μm depending on the As surface coverage during growth. When growth is initiated on the flat, singular ($\overline{1}\ \overline{1}\ \overline{1}$) surface, isolated pyramids are formed. As the growth proceeds, pyramids are generated over the entire surface until they start to overlap each other. Once the growth thickness has exceeded some value (which depends on d), the initially flat surface is completely covered by pyramids, and the structure remains stable on the growing film surface so long as the substrate temperature and the Ga/As$_2$ flux ration are held constant. Within the $\sqrt{19} \times \sqrt{19}$ reconstruction growth regime,
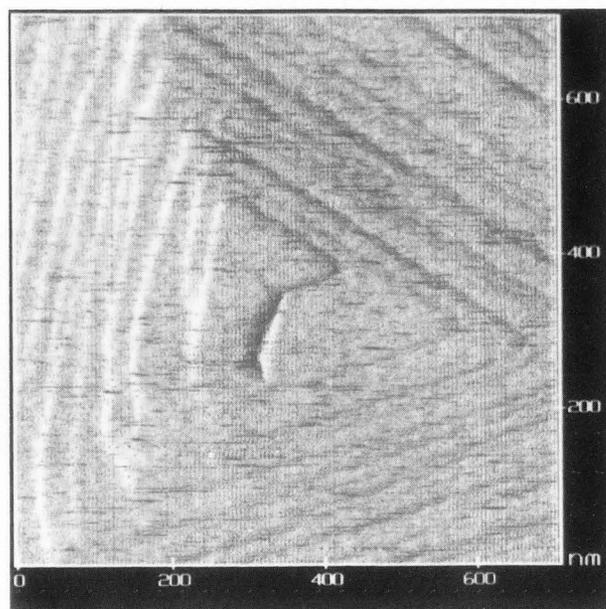
Figure 2. An atomic force microscope (AFM) image of the top of one of the pyramids shown in Figure 1. The scale is shown in nanometers.

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at the same fluxes, the pyramids were generated faster and the distances between pyramids were smaller at the lower substrate temperatures. The surface of a film grown in the low-temperature end of the $\sqrt{19} \times \sqrt{19}$ reconstruction regime (where d = 1 μm) was fully covered by pyramids after only 50 nm of deposition. These pyramids seem to remain stable even when the Ga flux is interrupted so long as the As_2 flux is adjusted to keep the surface in the $\sqrt{19} \times \sqrt{19}$ regime. When the surface is allowed to enter the 1x1 by either heating it to higher temperatures at constant As_2 flux or by reducing the As_2 flux at constant temperature, the pyramids rapidly disappear leaving a smooth surface.

In Figure 2, an AFM image is shown of the region near the top of an individual pyramid in which the atomic steps can be clearly seen. These steps should be understood to be a replica of the original, "clean" GaAs surface since the AFM images were taken in air. However, the step heights are very close to those expected for the (111) GaAs surface, and the average spacing between steps is approximately 7.5 nm which is what would be expected given the average slope of the vicinal surfaces of the pyramid. The steps are observed to run along the three $<0\,\overline{1}\,1>$ directions that lie in the surface plane. The "step-down" directions are along the $[2\overline{1}\,1]$, $[\overline{1}\,\overline{1}\,2]$ and the $[\overline{1}\,2\,\overline{1}]$ azimuthal directions. (i.e., if one crosses a step which runs along the $[0\,\overline{1}\,1]$ direction, one will step down in the $[2\,\overline{1}\,1]$ direction).

We have also investigated how the surface morphol-

ogy evolves during homoepitaxial growth in the $\sqrt{19} \times \sqrt{19}$ reconstruction regime when vicinal GaAs $(\overline{1}\,\overline{1}\,\overline{1})$ substrates of various miscuts are used. As we have shown in prior work (Yang *et al.*, 1993), homoepitaxial growth of GaAs on vicinal substrates, where the surface normal is tilted more than 3° toward the $[2\,\overline{1}\,\overline{1}]$ azimuthal direction, results in surfaces which appear to be very smooth when observed optically. Examination with the AFM of homoepitaxial layers on these substrates reveals an array of parallel atomic steps running along the $[0\,\overline{1}\,1]$ direction. These steps appear to fairly uniformly spaced which is consistent with the optical microscope observations of a very smooth surface.

A very different kind of surface morphology is observed when homoepitaxy on vicinal substrates tilted 1° or 2° toward the $[2\,\overline{1}\,\overline{1}]$ azimuthal direction as shown in Figures 3 through 6. In this situation, the surface morphology forms a grating-like structure. The grating consists of two facet orientations which are extended along the $[0\,\overline{1}\,1]$ direction. As the AFM height scan along the $[2\,\overline{1}\,\overline{1}]$ direction shows, the facets making up the grating are very nearly parallel to each other. Of course, the average orientation of the surface remains fixed at the original miscut of the substrate. Measurements of the angle between the two facets give a cluster of values at 2.7° ± 0.2° although occasional values (down to 1.9°) were observed. These smaller angles seemed to be more prevalent on samples which had a larger miscut (the 2° substrates) than on the vicinal samples with a smaller miscut. At higher resolution (an example of which is shown in Figure 5), we find that one of the facets has a low density of steps while the other facet has a high step density which corresponds to approximately a 2.5° vicinal surface. Note that the low step density facet for the 1° vicinal substrate is much wider than it is for the 2° substrate as one would expect given the requirement that the average orientation of the surface must be kept constant.

One should note that the results presented above on vicinal substrates are not what one would expect after observing the pyramid structure on the well-oriented substrates. One would predict rather that as one tilts toward the $[2\,\overline{1}\,\overline{1}]$, the pyramids would simply appear to be tilted until one reached 2.5° after which the surface would be smooth. Certainly, as the degree of miscut toward the $[2\,\overline{1}\,\overline{1}]$ is reduced from 3° to smaller angles, the formation of complete pyramids must occur at some point since we observe them on the singular $(\overline{1}\,\overline{1}\,\overline{1})$ substrates. Why do we not see tilted pyramids on the vicinal substrates when the angle of miscut is less that 3°? This question is partially answered by the observation of isolated pyramids on the 1° vicinal substrate such as the one shown in Figure 6. While the density of these pyramids is rather low on the 1° vicinal substrate,
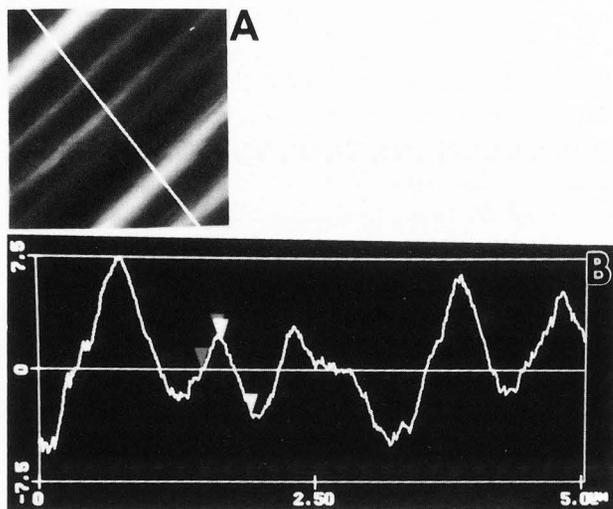
Figure 3. An AFM image of the surface morphology of a 1-μm-thick homoepitaxial film on a vicinal GaAs ($\overline{1}\ \overline{1}\ \overline{1}$) substrate which is tilted 2° toward the [2 $\overline{1}$ $\overline{1}$] azimuth. The line across (**A**) shows the path taken for the profile shown in (**B**). This film was grown while maintaining the $\sqrt{19}$ x $\sqrt{19}$ surface reconstruction. The growth parameters are described in more detail in the text.
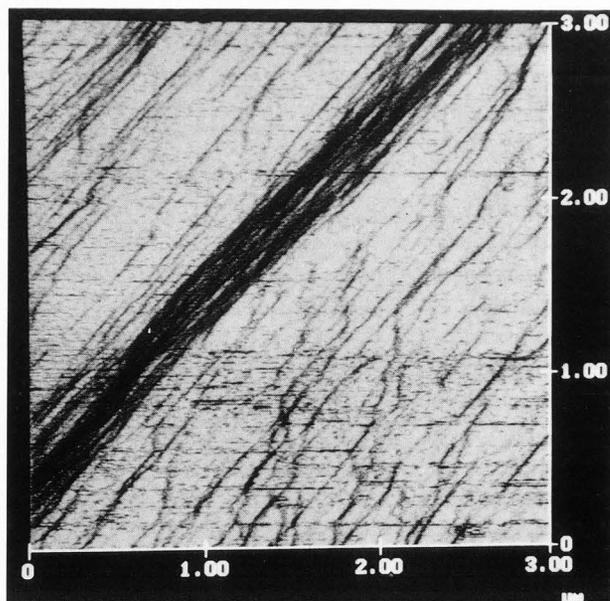

Figure 5. A higher resolution image of the sample shown in Figure 4 showing atomic steps (black lines) on the singular and vicinal facet. Note that the length scale here is measured in microns so that the atomic step density on the vicinal facet appears very dense (average spacing there is approximately 7.5 nm).
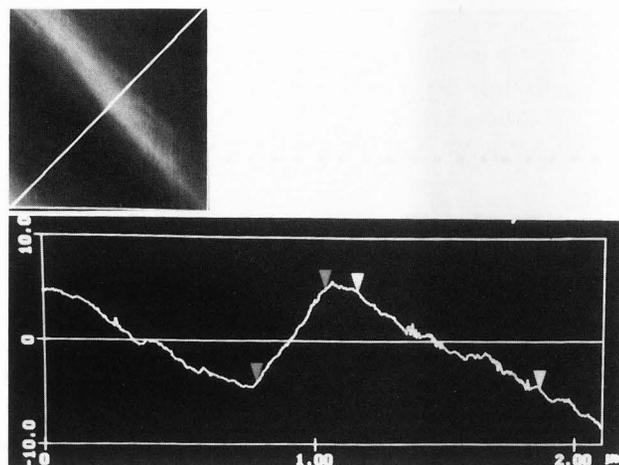

Figure 4. An AFM image of the surface morphology of a 1-μm-thick homoepitaxial film on a vicinal GaAs ($\overline{1}\ \overline{1}\ \overline{1}$) substrate which is tilted 1° toward the [2 $\overline{1}$ $\overline{1}$] azimuth. The growth conditions used were the same as for the sample shown in Figure 3.
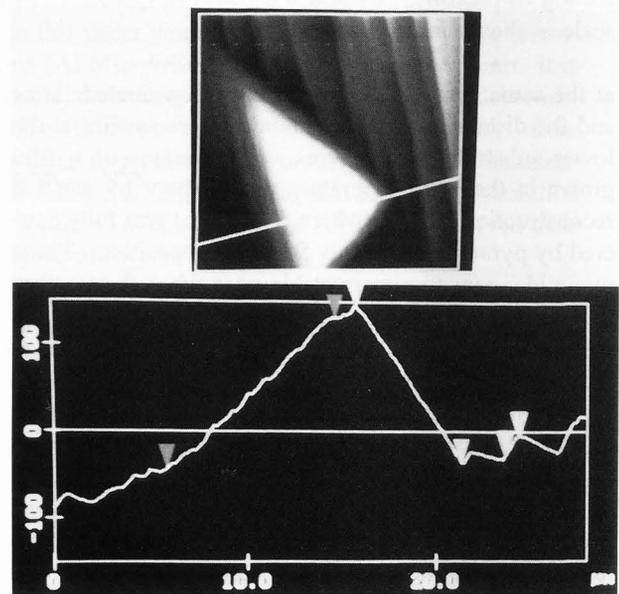

Figure 6. Another AFM image of the same sample shown in Figure 4 at a different place on the surface. Here a tilted pyramid has nucleated.

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we did not find any on the 2° substrate. It appears that the width of the singular substrate must exceed some value before pyramids structures can be nucleated.

RHEED Observations

Reflection high energy electron diffraction (RHEED) patterns also provide useful information about

Discussion

These results seem to be most consistent with the explanation that surface free energy of a tilted surface is less than that of the singular surface. Other possible explanations include the possibility that defects in the epitaxial layer control the formation of pyramids or that the Schwoebel effect causes the preferential formation of steps across the surface. We believe that we can effectively rule out the explanation that defects are controlling the nucleation of pyramids for several reasons. We can vary the distance between pyramids from 1 to 30 μm, but we see no change in the crystal quality as measured by RBS and with mobility measurements (Yang, 1993; Yang et al., 1993). In addition, the defect explanation would be inconsistent with the results we have obtained for vicinal substrates.

The Schwoebel effect refers to the energy barrier that a diffusing adatom sees when it approaches a step edge (Ehrlich and Hudda, 1966; Schwoebel and Shipsey, 1966; Schwoebel, 1969). Recently, this effect was used to explain large mounded features observed on the homoepitaxial surface of singular GaAs(100) substrates (Johnson et al., 1994). However, in the case of GaAs (001), the features are very irregular and do not show the very organized step structures that we observe for the 2.5° vicinal facets that form distinctive pyramids on the $(\bar{1}\ \bar{1}\ \bar{1})$ surface. In addition, homoepitaxial growth on the 1° and 2° vicinal substrates results in a faceted surface consisting of 2.5° vicinal surfaces and singular surfaces. The fact that the facet faces are parallel suggests that there is a thermodynamic driving force forcing a phase separation of the growing surface into 2.5° and singular regions. Our results suggest that the free energy of the singular regions is actually higher than that of the 2.5° vicinal regions. However, we continue to see singular regions until their width becomes large enough to nucleate the other two vicinal 2.5° surfaces whose surface normals are tilted in the $[\bar{1}\ 2\ \bar{1}]$ and the $[\bar{1}\ \bar{1}\ 2]$ azimuthal directions (as opposed to the $[2\ \bar{1}\ \bar{1}]$ direction).

We should note that we have not been able to achieve the same surface morphology simply by heating the GaAs$(\bar{1}\ \bar{1}\ \bar{1})$ substrate even when an appropriate As$_2$ beam is used to maintain the surface stoichiometry. This can be understood by the fact that the mobility of Ga is substantially greater during deposition. Recently, we (Yang et al., 1994) and others (Nomura et al., 1994) have shown that the diffusion length of Ga adatoms on the $\sqrt{19}$ x $\sqrt{19}$ surface must be at least several hundreds of nanometers. However, these conditions are difficult to duplicate under non-growth conditions. As described above, the pyramids will remain stable when the Ga flux is shut off so long as the As$_2$ flux is maintained
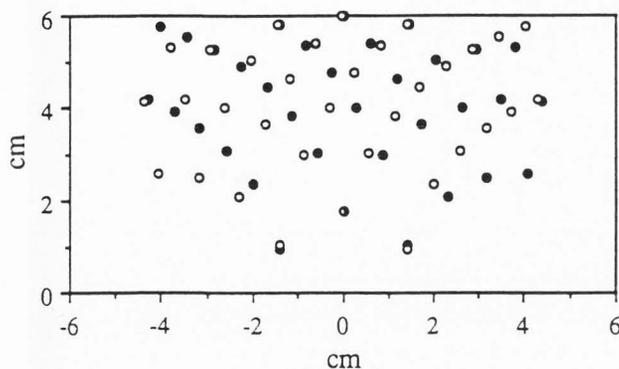

Figure 7. A calculated RHEED pattern along the $\langle 0\ 1\ \bar{1} \rangle$ azimuthal direction for the $\sqrt{19}$ x $\sqrt{19}$ reconstruction. The open circles are for $\sqrt{19}$ x $\sqrt{19}$ R+23.4° reconstruction, and the closed circles are for the $\sqrt{19}$ x $\sqrt{19}$ R-23.4°.

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the step structures. Figure 7 shows the expected RHEED diffraction spot positions for both $\sqrt{19}$ x $\sqrt{19}$ reconstructions when the electron beam is directed along a $\sqrt{19}$ x $\sqrt{19}$ azimuth (Yang, 1993). Figure 8A shows a typical RHEED pattern of the $\sqrt{19}$ x $\sqrt{19}$ reconstruction on a GaAs film grown on a singular $(\bar{1}\ \bar{1}\ \bar{1})$ substrate. Notice that sharp diffraction spots are observed, indicating long range ordering of the atomic steps . (These spots should not be confused with spots caused by transmission electron diffraction that can result in samples with much larger facet angles. Experimentally, it is easy to distinguish between the two since transmission electron diffraction spots will remain fixed in position as the substrate is rotated while RHEED spots will slide up or down on the screen as the corresponding reciprocal lattice rod cuts the Ewald sphere at different points.) We observe equal intensities of the two possible $\sqrt{19}$ x $\sqrt{19}$ reconstructions.

Figures 8B and 8C show RHEED patterns of the $\sqrt{19}$ x $\sqrt{19}$ reconstruction on a GaAs film grown on a vicinal $(\bar{1}\ \bar{1}\ \bar{1})$ substrate tilted 3° toward the $[2\ \bar{1}\ 1]$ azimuth. As we indicated in **Surface Structure**, films grown in this orientation will result in smooth surfaces which, when examined with an AFM, will only have parallel atomic steps running in the $[0\ 1\ \bar{1}]$ direction with an average spacing of about 6 nm. In Figure 8B, the electron beam is directed along the $[0\ 1\ \bar{1}]$ azimuth parallel to the atomic step edges while in Figure 8C, the beam is directed along the $[1\ 0\ \bar{1}]$ azimuth. Notice that the RHEED pattern in Figure 8B still shows sharp spots (and approximately equal intensities for the two possible $\sqrt{19}$ x $\sqrt{19}$ reconstructions) while in Figure 8C, the spots have elongated into streaks, indicating that the long range ordering of the atomic steps has been lost.

Figure 8. The RHEED pattern of the $\sqrt{19} \times \sqrt{19}$ reconstruction of: **(A)** a well-oriented GaAs surface along the [0 1 $\overline{1}$] azimuth; **(B)** along the [0 1 $\overline{1}$] azimuth of a vicinal substrate tilted 3° toward the [2 $\overline{1}$ $\overline{1}$] direction; and **(C)** along the [1 0 $\overline{1}$] azimuth on the same substrate (in this last case, the electron beam makes an angle of 60° to the step edges). Note that the sharp spots observed in (A) and (B) have evolved into streaks in (C).

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to keep the surface reconstruction in the $\sqrt{19} \times \sqrt{19}$ regime. If the substrate surface is allowed to anneal in the 1x1 reconstruction regime, the pyramids rapidly disappear. These results suggest that the formation of the vicinal surfaces is thermodynamically controlled (i.e., they have a lower free energy than the singular surface).

It is generally believed that crystal surfaces which are exactly parallel to a low-index Miller plane should have a lower free energy than a vicinal surface consisting of exactly oriented terraces separated by atomic steps. However, Alerhand *et al.* (1988, 1990) have pointed out a mechanism for vicinal surfaces to have a lower free energy than an exactly aligned (singular) crystal surface if the surface reconstruction has two degenerate reconstructions which cause anisotropic surface stresses. In our case of the $\sqrt{19} \times \sqrt{19}$ reconstruction, the two degenerate reconstructions are rotated $\pm 23°$ with respect to the unreconstructed bulk, resulting in different torques and, thus, anisotropic stresses when terminated at a step edge. Alerhand *et al.* (1988, 1990) and others (Tersoff and Pehlke, 1993) have applied this model to the 2x1 Si(001) surface. While the situation there is different in several fundamental ways (for instance, single atomic steps rotate by 90° the orientation of the reconstruction), the general argument by Tersoff and Pehlke (1993) showing that the surface free energy will have a minimum at a vicinal angle greater than 0° away from the singular surface should also be valid here. As shown by Williams *et al.* (1993), this will lead the surface to facet if it can achieve its equilibrium configuration. We believe the low step density surfaces which are observed on the 1° and 2° vicinal surfaces result because the facets are too narrow to nucleate the lower energy surfaces. As the width of the nearly singular facets are increased, pyramid structures are nucleated.

It should be noted that the mechanism proposed here is quite different than that proposed for the faceting that is observed on Si(111) surfaces. In that case, the singular surface exhibits a surface reconstruction while the vicinal facets have the 1x1 high-temperature reconstruction. Both of these reconstructions would have a minimum in their surface free energy at the singular surface ($\theta = 0$), however, they have different dependencies on θ which results in a first-order phase transition (Williams
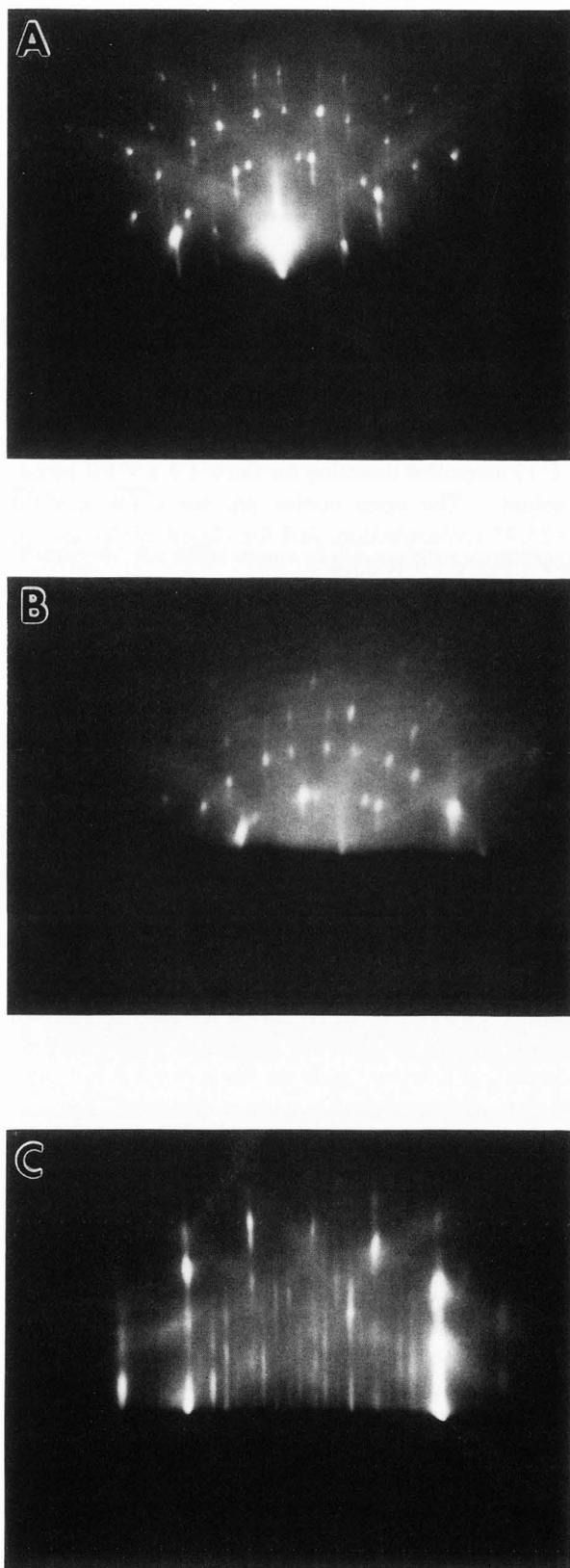
et al., 1993). These different mechanisms point out the richness of surface morphologies possible under different growth conditions and with different materials systems.

Conclusions

We have observed that under homoepitaxial growth in the $\sqrt{19}$ x $\sqrt{19}$-surface-reconstruction regime, the singular $(\bar{1}\ \bar{1}\ \bar{1})$ surface of GaAs spontaneously breaks up into vicinal surfaces which are approximately tilted 2.5° toward the three equivalent $<2\ \bar{1}\ \bar{1}>$ azimuthal directions (keeping in mind that the $[2\ \bar{1}\ \bar{1}]$ and $[\bar{2}\ 1\ 1]$ directions are not equivalent). This results in the formation of three-fold symmetric pyramids. If vicinal substrates, with a tilt greater or equal to 3° toward the $[\bar{2}\ 1\ 1]$ are used, very smooth surfaces can be grown where no atomic step bunching is observed. Growth on vicinal substrates with smaller angles of tilt will result in faceting where one set of facets is singular (low step density) and the other set of facets are tilted approximately 2.5° toward the $[\bar{2}\ 1\ 1]$ azimuth. We believe these results can best be understood as caused by the 2.5° vicinal surface having a surface-free-energy minimum. This minimum could be explained as the result of a surface anisotropic strain due to the degenerate $\sqrt{19}$ x $\sqrt{19}$ reconstructions that are possible on this surface. We also observed that the singular facet must be at least 1 μm wide before the vicinal surfaces can be nucleated.

These results allow a more complete understanding of the surface morphologies than have been observed by other groups working on GaAs$(\bar{1}\ \bar{1}\ \bar{1})$ substrates. Low temperature growth of smooth surfaces on vicinal $(\bar{1}\ \bar{1}\ \bar{1})$ substrates can be achieved when the substrate is appropriately tilted toward the $[2\ \bar{1}\ \bar{1}]$ azimuth. Thus, high quality multilayer structures of In$_x$Ga$_{1-x}$As are possible. We also expect that the high degree of step organization that is observed on this surface could be utilized to grow quantum wire and quantum dot structures. Finally, our results demonstrate another possible mechanism for introducing atomic-step organization in growth on crystal surfaces which are closely oriented to high symmetry directions.

Acknowledgments

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Discussion with Reviewers

B. Orr: Is there any way of predicting the vicinal angle of the surface which is thermodynamically preferred? In other words, is there a simple geometric scheme of tilting the $\sqrt{19}\times\sqrt{19}$ reconstructions to see why the 2.5° (7.5 nm terraces) vicinal surface has a lower energy?
Authors: One possibility would be that the terraces would be a "magic" integral number of $\sqrt{19}\times\sqrt{19}$ unit cells. However, the terraces we observe seem to be too large for that possibility. We think that it is more likely that the distance between steps is explained by a competition between energy advantage of introducing an individual step versus the cost in energy of steps interacting with each other (i.e., step-step repulsion).

Reviewer I: One of the main claims of the paper is the identification of the 2.5° vicinal surface as the energetically preferred surface. Such a claim is internally inconsistent with the authors' own observations on 1° and 2° vicinal substrates. I fail to see why the existence of the pyramids should depend on the size of the terraces, if thermodynamics is the driving force for the observed structures.
Authors: Of course, there are many situations where a critical size is needed to nucleate a new phase. For instance, the surface energy of water causes water nuclei below some critical size to be unstable. In the present work, a similar situation exists with the tops of the pyramids where the atomic steps cannot be distributed in the same way that they along the faces of the pyramids. However, the reviewer makes a good point that we cannot, with the data we have, distinguish between a true minimum in the free energy at 2.5° versus a local minimum. This issue is currently unresolved.

Reviewer I: All the data shown are for very high coverage growth (1 μm). At such coverage, contamination is a serious concern. I have difficulty seeing why such a coverage is needed for the pyramids to cover the surface, if the energetics were indeed the driving force. From what is presented in the paper, I do not think the possibility of contamination can be ruled out.
Authors: This concern about contamination seems totally inappropriate. Why would contamination be more of

a problem for thicker layers? In addition, as stated in the paper, we do see the pyramids forming from the very start of deposition when the surface is kept in the $\sqrt{19}\times\sqrt{19}$ reconstruction during deposition.

Reviewer I: From a theoretical point of view, I do not see how the argument used for Si(100) can be used here. The (111) surface has 3-fold symmetry and the $\sqrt{19}\times\sqrt{19}$ reconstruction preserves such symmetry. As a result of such high symmetry, the surface stress is isotropic. Thus, there is no mechanism for the surface to lower its energy by creating steps.
Authors: We agree that the $\sqrt{19}\times\sqrt{19}$ reconstruction preserves the 3-fold symmetry of the (111) surface. However, this three-fold symmetry is broken once steps are introduced. If the surface reconstruction is ignored, the three-fold symmetry can be preserved when steps are introduced by running the steps along the three symmetry directions. However, this is no longer possible when the surface reconstructs in a particular $\sqrt{19}\times\sqrt{19}$ reconstruction which is rotated $\pm23°$.