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SUPPRESSION OF ANTIPHASE DOMAINS IN THE GROWTH OF GaAs ON Ge(100) BY MOLECULAR BEAM EPITAXY

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We have used reflection high energy electron diffraction (RHEED) to determine the step structure on Ge and GaAs surfaces. The results show the key role that inequivalent steps on Ge substrates play in suppressing antiphase domain formation at the GaAs/Ge(100) interface. For Ge surfaces misoriented toward the [011] direction and exposed to an As flux, a mass migration causing a four-monolayer step periodicity is observed. If a Ga flux is then applied, a single domain of GaAs is formed. The single domain is observed at the start of growth with weak half order streaks appearing in only one azimuth. In contrast, for surfaces misoriented exactly toward the [010]. RHEED measurements show a different distribution of multilayer step heights with no clearly predominant even or odd layer step height. GaAs grown on this surface shows a reconstruction consisting of two domains.

1. Introduction

The recent successful efforts to grow high quality GaAs on both Ge and Si by molecular beam epitaxy (MBE) [1–7] immediately lead to questions concerning the microscopic processes of the hetero-epitaxy. For either Si or Ge substrates one needs to determine the growth mode in which antiphase domains are avoided. These antiphase domains have long been an issue [7,8] and are an expected consequence of the growth of zincblende on diamond. For growth on Si substrates one would also like to understand the full impact of the severe lattice mismatch. In the following we present a reflection high-energy electron diffraction (RHEED) study of the growth of GaAs at the less complicated, GaAs/Ge(100) interface. Strain should be much less important in this nearly lattice matched system. The results indicate that the initial step distribution of the substrate is crucial to the subsequent growth mode and that the step distribution is changed by reaction with an As ambient.

The formation of antiphase domains (APD) is closely connected with the surface morphology and is a consequence of the equivalence of the two sublattices of the diamond structure. The [100] direction in Ge consists of alternating planes of Ge atoms in which the bonding is rotated by $\pi/2$. In GaAs not only is the bonding rotated but also the alternating planes are in ABAB stacked sequence of Ga and As. For growth on an ideal, flat (100) surface, antiphase domains would be avoided if the first layer is forced to be either entirely Ga or entirely As. One attempts to accomplish this by adsorbing As before growth and then slowly initiating the growth of GaAs in an As rich environment [2]. However, if there are monolayer steps on the Ge surface, i.e. steps of height equal to a/4 where a is the lattice constant (see fig. 1), both sublattices will be exposed and saturating the first layer will still lead to APD's.

We will use RHEED to measure the distribution of steps on the Ge surface prior to GaAs

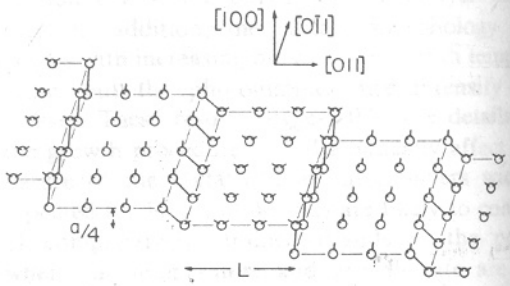

Fig. 1. Perspective view of a Ge(100) surface showing single layer steps and change in exposed sublattice after a step. The misorientation is toward the [011] direction and the lattice constant, a, is 5.65 Å.

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growth and then to detect the presence of APD's at the GaAs surface. For the first measurement the shape of the specular beam is determined as a function of scattering angle. The beam will be broad or split depending on the regularity of the terrace lengths and step heights [9]. This is a consequence of constructive and destructive interference between waves scattered from different terraces. By measuring the diffracted intensity along the length of the specular beam (streak), we will show that when the Ge surface is misoriented in the [011] direction, either As_4 or As_2 adsorption causes a mass migration that produces a staircase with periodicity equal to the lattice constant, a, and not a surface with simple double-layer step heights. GaAs subsequently grown on this surface contains only a single domain. In contrast, when the crystal misorientation is in the [010] direction we will show that an ordered array of steps is not obtained and that in this case the APD's are kinetically, but not energetically allowed.

2. Experimental

The diffraction and growth apparatus have been described elsewhere [10]. The growth was performed in a Perkin-Elmer MBE 400 system. The Ge surfaces were prepared by growing a 1000 Å layer of Ge onto the misoriented GaAs substrates listed in table 1. The Ge layer was grown at a rate of 0.2 Å/s. A PBN crucible was used to deposit the Ge so that the background pressure during growth was 5×10^{-9} Torr [4]. The subsequent GaAs layer was grown with either As_4 or As_2. The flux was set to give an As incorporation rate of 1.5 monolayers/s at a substrate temperature of 500°C based on RHEED As oscillations [11]. Initially the growth temperature was 500°C and growth rate 0.1 μm/h. After 200 layers were deposited, these were increased to 580°C and 0.8 μm/h respectively. The final GaAs thickness was 2 μm in all the films.

Table 1
Step and domain structures observed on Ge(100) misoriented substrates; GaAs domain orientation, $[01\bar{1}]$, indicates the direction of the two-fold reconstruction

Substrate misorientation		As species	Four-layer step period?	GaAs domain
θ (deg)	ϕ			
6	[011]	As_4	Yes	$[01\bar{1}] \perp$ to ϕ
6	[011]	As_2	Yes	$[01\bar{1}] \parallel$ to ϕ
2	[011]	As_4	Yes	$[01\bar{1}] \perp$ to ϕ
2	[010]	As_4	No	Two domains
0.25	[041]	As_4	No	$[01\bar{1}]$ 69° to ϕ + weaker domain $[01\bar{1}]$ 31° to ϕ

3. Results

The most defect free growth of GaAs on Ge has been observed on substrates misoriented by a few degrees from the (100) [2]. We have followed this approach and used substrates misoriented toward the [011] and [010] directions. In the following section we report our characterization of the surface during each step leading to single domain growth of GaAs on Ge(100). Our Ge results are similar to those reported by Olshanetsky and coworkers [14] for the [011] direction, but out interpretation is very different. The change in the surface after As adsorption shows that initial stages of growth are more complicated than had previously been thought.

3.1. Ge(100) surfaces

After growing the Ge film and raising the temperature to 500°C, an ordered array of steps on the misoriented surfaces was confirmed by RHEED, For a 2° [011] surface, this is shown in fig. 2a where the diffracted intensity along the length of the specular streak exhibits characteristic sharp splitting. The electron beam is 10° off the [011] direction. There are two important issues to be noted. First for this ordered staircase, we always observe orthogonal 2×1 and 1×2 domains [12]. The fractional order beams are of nearly equal intensity so that neither domain is dominant. It is not a 2×2 reconstruction since the $(\frac{1}{2}\frac{1}{2})$ beam is missing at all scattering geometries. This means that the staircase contains significant numbers of terraces of both sublattices and hence

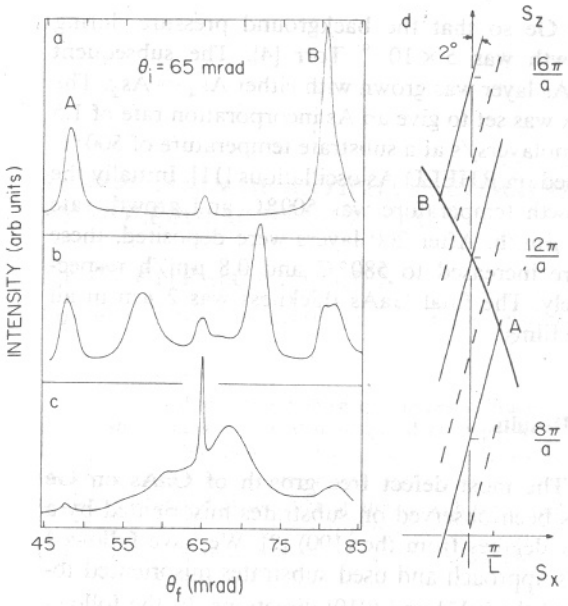

Fig. 2. (a) RHEED specular beam splitting from Ge(100) 2° [011] surface. Profile is measured along the length of a streak. Locations of the more intense, outer peaks indicate single-layer step heights. The incident angle is 65 mrad with the 10 keV beam directed in a [011] azimuth (down the staircase step array). (b) Diffracted beam profile from Ge(100) 2° [011] surface after exposure to As flux. New peaks at 57 and 73 mrad indicate a step periodicity of four layers or one lattice constant, a, high. (c) Diffracted beam profile from Ge(100) 2° [010] surface after exposure to As flux. The lack of distinct peaks about the specular position indicate a surface composed of a range in step heights. Prior to As exposure, the profile appeared similar to that in (a). (d) Ewald sphere intersection with reciprocal lattice of a 2° surface. Reciprocal lattice lines were calculated assuming a staircase of steps. Intersection points A, B correspond to A, B in (a). Dashed lines of reciprocal lattice only appear if step periodicity is 4 monolayers. Average terrace length, $2L$, is $a/\tan 2° = 160$ Å.

some single-layer steps of height $a/4$. Second, as explained below, the scan of the intensity along the specular streak shown in fig. 2a indicates that single-layer steps dominate. A similar single-layer periodicity is observed for the other misorientations. We disagree with the interpretation of Olshanetsky and coworkers that surfaces misoriented toward the [011] contain primarily double-layer steps.

The separation of the two intense peaks, labelled A and B in fig. 2a, corresponds to diffraction from a surface with single-layer steps. The Ewald construction with the same labelling is shown in the panel on the right. The weak central peak corresponds to a less pronounced $a/2$ periodicity which is also present (larger periodicities give a smaller spitting). If double-layer steps dominated, the central peak would be stronger. Two effects besides double-layer steps could give the small central peak: (1) the different domains could have different scattering factors and (2) the lengths of the terraces could alternate [13,17]. Both would give an $a/2$ periodicity. Because similar relative intensities are measured at other scattering geometries, it is impossible to fit the data without modelling the surface staircase with primarily single-layer steps.

3.2. Ge(100): As surfaces

A major reorganization of the Ge step structure occurs on each of the misoriented surfaces if exposed to an As flux. This is indicated by further splitting of the specular beam, illustrated in fig. 2b, that results from a longer surface periodicity. The position of the new peaks as a function of scattering angle for both 2° [011] and 6° [011] surfaces correspond to a staircase structure that repeats after four layers. This four-layer periodicity gives additional constructive interference conditions which are halfway between the peaks shown in fig. 2a. The extra reciprocal lattice rods are shown in fig. 2d as dashed lines. During this step redistribution, the positions of the fractional-order peaks were unchanged.

The transition from single-layer steps to a four-layer periodicity is reversible as shown in fig. 3. At high As fluxes and low substrate temperatures the four-layer structure dominates. The phase boundary has a latent heat of 5 eV, signifying a first-order phase transition. The width of the transition region is $\leq 10°C$. Though the phase boundary shown in fig. 3 was determined with an incident As flux, the transition could be initiated by adsorbing a small fraction of a monolayer of As, suggesting that adsorption on the risers of the original steps is responsible for the transition. At low temperatures, the four-layer period is stable for a long time without an As flux. After heating

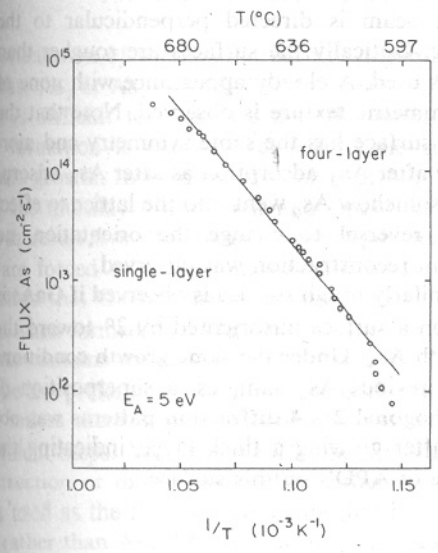

Fig. 3. Phase boundary between the Ge(100) 2° [011] single-layer and As induced four-layer structure. As_4 used as the incident flux with calibration explained in the text. The sticking coefficient is assumed to be the same as for GaAs. The latent heat, E_A, is 5 eV.

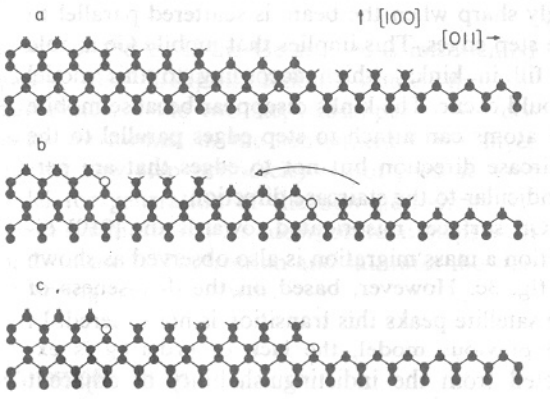

Fig. 4. (a) Ge(100) single-layer stepped surface. (b) As adsorption at step edges (open circles) leads to mobile adlayer of Ge. 9c) Complete transition to four-layer periodicity (single-layer step + three-layer step).

to regain the single-layer structure, As could not be detected by Auger spectroscopy.

Since we still observe two orthogonal 1×2 domains with near equal intensity on the As covered Ge surface, even-multiple step heights alone cannot explain the diffraction data. Simply attributing the observed four-layer periodicity to four-layer high steps would not lead to two domains. There must be some odd-multiple step heights. The exact shape of the diffraction profile also leads to a more complicated picture. If the surface were composed of only four-layer steps, the central peak of fig. 2b would be of greater intensity than the satellite peaks. Instead the central peak is one fourth the intensity of the neighbors. This is consistent with data at other scattering geometries, indicating a large proportion of odd-layer step heights.

A simple model that gives a two domain reconstruction and a four-layer periodicity is one that alternates triple- and single-layer steps. Assuming that the scattering factor for both domains are nearly identical, the calculated diffraction profiles from this surface are very close to what is measured in fig. 2b. Though not unique, it is one of the simplest reasonable models that could be presented. One way to form this surface requires (1) saturable reaction of steps containing bonds in the [111] direction and (2) a high mobility of surface Ge atoms. Fig. 4 illustrates the model. Fig. 4a is the Ge(100) vicinal surface consisting of single-layer steps. Arsenic atoms are allowed to replace Ge atoms only at those step edges shown in fig. 4b. When this reaction takes place, the Ge atoms located to the right of the As sites are more weakly bound due to elimination of the corner bond that results from threefold coordination of the substitutional As [15]. The remaining Ge on this terrace migrate over the surface to a lower energy configuration. They are not allowed to attach to a step with substitutional As bonding at the corner. The arrow in fig. 4c illustrates one path that allows the weakly bound islands to coalesce and form an alternating triple- and single-layer step periodicity. From RHEED studies of ordering rates from deliberately roughened surfaces [13], a substrate temperature of $> 500°C$ is high enough to allow an average adatom migration of length 40 Å to proceed quickly. There are two results that corroborate this mechanism. First, after the As is adsorbed, Ge will not grow epitaxially, suggesting that the step propagation growth mechanism is less effective. Second, after As is adsorbed, the diffracted beams become exceed-

ingly sharp when the beam is scattered parallel to the step edges. This implies that mobile Ge is able to fill in kinks, which according to this model should occur. The kinks disappear because mobile Ge atoms can attach to step edges parallel to the staircase direction but not to edges that are perpendicular to the staircase direction.

On surfaces misoriented towards the [010] direction a mass migration is also observed as shown in fig. 3c. However, based on the diffuseness of the satellite peaks this transition is not ordered. In the previous model, the lack of ordering is expected from the indistinguishability of adjacent terraces and their step edge sites to As adsorption.

3.3. GaAs(100) / Ge(100)

The growth of GaAs on Ge initially follows a three-dimensional process [1,2]. Initiating growth with an As_4 flux on a surface misoriented toward the [011], we have observed well resolved chevrons superimposed on the bulk spots when the electron beam is directed perpendicular to the staircase. The angle of the chevrons with respect to the [100] direction indicates that the bulk diffraction is occurring through (311) facets. Within 1000 Å of growth, these chevrons fade and 1/4 order streaks are observed, establishing the facets as (311)B. With the electron beam directed down the staircase, weak 1/2 order streaks superimposed on a transmission-like pattern are observed quickly after the start of growth implying an immediate orientation of the overgrowth and that the asperities are long in the [01$\bar{1}$] direction. Further growth on such a surface results in split diffraction spots characteristic of a misoriented GaAs surface with only the usual bilayer As–As steps. Optically the film shows a texturing on the surface with the lines in the [01$\bar{1}$] direction.

The crucial observation is that immediately after only one equivalent monolayer of GaAs is deposited, a superposition of Ge and GaAs patterns is observed. Even though the substrate consists of both types of sublattices in near equal numbers, only one domain of GaAs is seen.

If As_2 is used to initiate growth, the GaAs domain observed is rotated 90° from the previous case, i.e. 1/2 order streaks are observed if the electron beam is directed perpendicular to the staircase. Optically the surfaces are rougher than if As_4 is used. A cloudy appearance with none of the asymmetric texture is observed. Note that the starting surface has the same symmetry and morphology after As_2 adsorption as after As_4 adsorption. If somehow As_2 went into the lattice to effect a bond reversal to change the orientation, no change in reconstruction was observed.

A similarly rough surface is observed if GaAs is grown on a surface misoriented by 2° toward the [010] with As_4. Under the same growth conditions as the previous As_4 samples, a superposition of two orthogonal 2×4 diffraction patterns was obtained after growing a thick layer, indicating the presence of APD's on this surface.

4. Discussion

Table 1 summarizes the results on five different surfaces. The main requirement for single domain growth of GaAs is that the misorientation be in a direction somewhat away from the [010]. A similar pattern has been observed for the growth of GaAs on a lens shaped Si(100) substrate [19]. When the misorientation is exactly in the [010] direction the bonding at a step from either sublattice is indistinguishable. A step from sublattice A to B is just the mirror reflection of a step from B to A. With no reason to prefer one domain over another, two domains of GaAs are observed. When the misorientation is directed away from the [010] two types of steps become available. For these misorientations, single domain GaAs can be grown.

From the diffraction measurements in two orthogonal directions, the initial GaAs growth cluster takes the shape of a truncated structure with a rectangular base and (311) facets perpendicular to the long direction. Similar facetting is observed in ref. [2]. Facet development may be the result of the difficulty of nucleating layer-by-layer with a polar material. Though the (311) face is polar, no charge accumulation or dipole is associated with the (311) pyramidal facet [16]. Further, the (311) is the slowest growth direction of Si [18]. One might suspect that the facets aligned with the multilayer steps on the substrate. But this is not correct since

the orientation of the facet structures depends on whether As_2 or As_4 is used for the growth.

It is difficult to conclude that the multilayer step formation is causally related to the single domain growth. Instead we can only conclude that both the multilayer transition and single domain growth result from the same step inequivalences. We are forced to this result because of the observation that GaAs clusters are immediately oriented when formed on a Ge(100) : As surface. This surface contains terraces of both sublattices. There can be no preference for either sublattice at the start unless there is preferred bonding at the inequivalent steps. We see no difference in the reconstruction or morphology when either form of As is used as the flux. We conclude that by using As_2 rather than As_4 the preference for nucleation at the different steps is reversed.

Once the domains are initiated we have evidence that energetics becomes more important. For a misorientation of 0.25° toward a [041] direction (15° from a [010]), a two domain 2×4 reconstruction is observed during growth at 580°C with As_4. After raising the temperature to 650°C, following Wang [5], one of the orthogonal domains becomes predominant, with the $[01\bar{1}]$ domain direction having a stronger component perpendicular to the staircase direction as expected from table 1. Raising the temperature allows the GaAs domain which initially is more prevalent to dominate, thereby eliminating energetically unfavorable APD's.

5. Conclusion

In summary, the misorientation and step distribution of Ge(100) surfaces have important consequences on the single domain growth of GaAs. We have used RHEED to study how the growth can be optimized to form a single domain. We have demonstrated that for Ge(100) surfaces misoriented toward the [011] direction, a quadrupling of the monolayer step periodicity occurs under an As flux. We argue that the inequivalence of the two possible steps between the two sublattices on a Ge surface are responsible for the formation of this new periodicity as well as single domain growth of GaAs. If the Ge surface is misoriented toward the [010] direction, where the inequivalence of the two steps disappears, two domains are formed in the subsequent GaAs layer and the new step periodicity is not present. Our model for generation of the new periodicity assumed substitutional As incorporate at steps with bonds in the [111] direction and saturates the step preventing further growth.

Acknowledgements

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