

## **Island Coalescence Induced Substructure Within GaP Epitaxial Layers Grown on (001), (111), (110) and (113) Si**

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### **ABSTRACT**

GaP islands grown on selected surfaces of Si and their coalescence behavior have been investigated by transmission electron microscopy. These layers were grown by chemical beam epitaxy. A number of significant observations emerge from this study. First, planar defect formation has been shown to be related to stacking errors on the smaller P-terminated {111} facets of GaP islands. Amongst the four orientations, (111) epilayers have a higher density of stacking faults and first order twins because of more P-terminated {111} facets per island. Second, multiple twinning on exposed {111} facets can produce tilt boundaries and irregular growths when islands coalesce. Third, inversion domain boundaries lying on {110} planes have been shown to form during GaP island coalescence across monatomic steps on (001) Si. Image simulations have been performed to show that these boundaries can be seen in high resolution lattice images and the observed contrast is attributed to the presence of wrong Ga-Ga and P-P bonds at the inversion boundary.

### **INTRODUCTION**

GaP and Si have a lattice mismatch of 0.37% at room temperature. Therefore, the two materials constitute an ideal system to study the effects of substrate orientation on the growth morphology and defect microstructure during polar on non-polar heteroepitaxy. Previously, we showed that GaP nucleated as faceted three-dimensional (3-D) islands on the (001) and (111) Si surfaces, while a nearly two dimensional (2-D) growth mode was seen on the (110) and (113) surfaces [1]. Further, it was observed that stacking faults and twins can be formed by stacking errors during growth on the smaller P-terminated {111} facets of 3-D epitaxial islands [2]. The observation of a more uniform 2-D growth mode for the (110) and (113) surfaces suggested that growth on these two Si surfaces may provide a plausible way of reducing the planar defect density in the coalesced GaP epitaxial films grown on Si. In this paper, we have examined the microstructure of coalesced growths on the aforementioned Si surfaces to delineate the effect of island coalescence on the defect density within the GaP epitaxial films.

## EXPERIMENT

The Si wafers were cleaned using an RCA clean. This consisted of a ten minute dip in a 1:1:5 solution of  $\text{NH}_4\text{OH}$ ,  $\text{H}_2\text{O}_2$  and DI water maintained at  $75^\circ\text{C}$ , a five minute rinse in DI water, a ten minute dip in a 1:1:5 solution of  $\text{HCl}$ ,  $\text{H}_2\text{O}_2$  and DI water also kept at  $75^\circ\text{C}$  and a rinse in DI water. This was followed by a buffered HF etch and a final DI water rinse. GaP was deposited on Si using pulsed chemical beam epitaxy (CBE) wherein, the heated Si substrate was exposed to pulses of triethylgallium (TEG) and continuous tertiarybutylphosphine (TBP) with a steady hydrogen background pressure [3]. The temperature range investigated was  $420\text{-}560^\circ\text{C}$  and the overall pressure during deposition was between  $10^{-4}$  and  $10^{-5}$  Torr. Structural analysis of as-grown layers was performed in cross-section by high resolution transmission electron microscopy (HRTEM) on a JEM-4000EX microscope operating at 400 kV and with an interpretable resolution of 0.17 nm.

## RESULTS AND DISCUSSION

Figs. 1 (a) and (b) show HRTEM images of (001) and (111) GaP epilayers grown at  $560^\circ\text{C}$  and  $420^\circ\text{C}$  for 500s and 300s, respectively. Stacking faults (SFs) and twins on  $\{111\}$  planes are the dominant feature of these epilayers. Stacking faults (SFs) are observed to intersect with other SFs ( $S_4$  and  $S_5$ ,  $S_6$  and  $S_7$ ) and intersect with microtwins ( $S_2$ ). Fig. 1 (b) shows a higher density of faults and twins parallel to the interface. The image also indicates that multiple twinning on island facets CD and FG can produce a tilt boundary  $B_1$  after island coalescence. The rough morphology of the epilayer surface further suggests the coalescence of faceted 3-D islands. Figs. 1 (c) and (d) are HRTEM interfacial images of thick (113) and (110) GaP epilayers grown at  $420^\circ\text{C}$ . For the (113) epilayers, a higher density of faults is present on the shallowly inclined  $\{111\}$  planes, while for the (110) epilayers faults are observed on both the shallowly inclined  $\{111\}$  planes. Continuously faulted regions with the anomalous ...AaXxYyAa.. are also observed within all four layers. In addition, irregular growth (IR) areas are seen in Figs. 1 (c) and (d) for (113) and (110) epilayers and in Fig. 2 (a) for (111) epilayers. A digital diffractogram (DDF) obtained from the irregular growth region marked as 1 in Fig. 2 (a) also shows satellite spots similar to the selected area diffraction pattern (SADP) in Fig. 1 (d).

The stacking fault and twin density is lower within (001) layers as compared to (111) GaP layers and within (113) epilayers as compared to (110) epilayers. Islands grown on the (111) surface are hexagonal in shape [1] and have 3-P terminated  $\{111\}$  facets per island, while islands grown on the (001) surface have 2-P terminated  $\{111\}$  facets per island. In addition, since P forms stronger bonds to Si [4,5], the Si surface will always be P-terminated. Thus the more complex shape of islands compounded with a higher number of P-terminated  $\{111\}$  facets (4 vs. 2) may be attributed to the higher defect density within (111) epilayers. For the (110) surface, two of the four  $\{111\}$  planes are shallowly inclined to the (110) surface ( $35.24^\circ$ ) and two of the planes are inclined  $90^\circ$  to the surface. The shallow facets  $(1\bar{1}1)$  and  $(1\bar{1}\bar{1})$  are terminated by P and Ga atoms, respectively. Low magnification images [6] have revealed a higher propensity for faulting on one of the two shallowly inclined  $\{111\}$  planes implying that this may be P-terminated. In the case of the (113) surface, only one out of the four  $\{111\}$  planes has a shallow inclination. Islands (even near 2-D ones as shown for these two orientations in [1]) would tend to facet along shallowly inclined  $\{111\}$  facets. Anomalous growth regions are

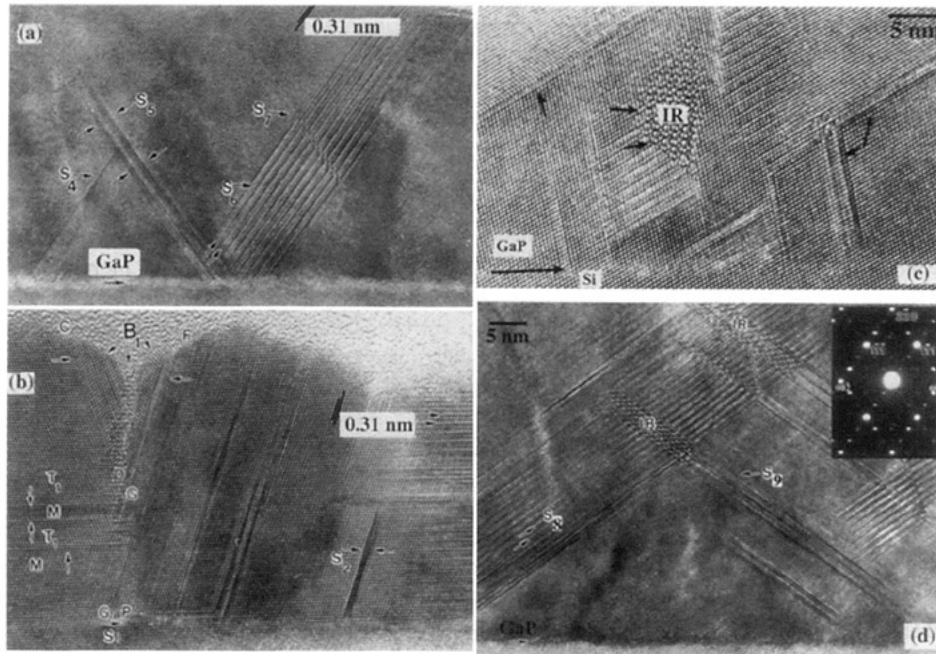


Fig. 1. (a), (b), (c) and (d) are HRTEM images of GaP epilayers grown on (001), (111), (113) and (110) surfaces of Si after 500s, 300s, 2300s and 2200s of growth. The (111), (110) and (113) Si epilayers were grown at 420°C and the (001) Si epilayer was grown at 560°C.

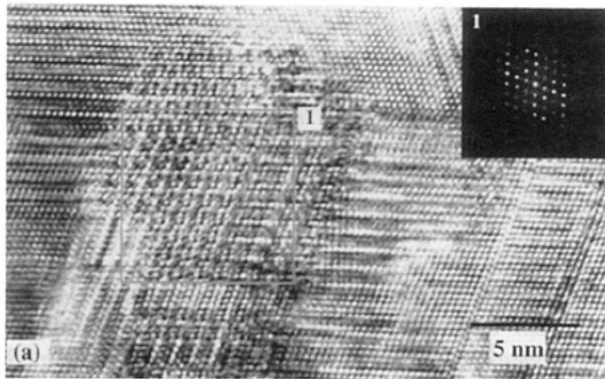


Fig. 2. (a) HRTEM image of a (111) GaP epilayer after 300s of growth at 420°C. The insert is a digital diffractogram obtained from the irregular growth region. (b) [110] simulated diffraction pattern indicating the additional spots due to twinning.

observed mostly on  $\{111\}$  planes which are shallowly inclined to the interface which implies that shallow facets provide a better template for faulting. This observation can be rationalized in terms of the larger projected areas of  $\{111\}$  facets exposed to incoming adatoms. Thus, faulting though possible on the steep facet appears to be more localized and less ubiquitous. Since there are a larger number of  $\{111\}$  facets at shallow incidence for faulting in the case of growth on (110) Si as compared to (113) Si, it is envisaged that these epilayers would exhibit a higher planar defect density.

The presence of anomalous growth sequences is not yet understood but it can be envisaged that the low growth temperatures impose kinetic restrictions (low mobility for instance) similar to the wurtzite stacking on certain  $\{111\}$  planes observed within GaP islands grown on (111) and (001) Si [1, 2]. A common feature within all four epilayers is the observation of IR at the intersection of the continuously faulted segments. We attribute the formation of these areas to multiple twinning on inclined  $\{111\}$  planes. We have indexed the SADP obtained from the (110) epilayers (which show the highest density of IR regions) assuming the twin plane is  $(1\bar{1}1)$  (Fig. 2 (b)). All the spots formed by the twin transformation appear as bright spots in the SADP. The presence of faint spots is attributed to double diffraction. The DDF obtained from an IR area in Fig. 2 (a) show additional spots similar to the SADP confirming our earlier supposition. Indeed, multiple twinning on inequivalent  $\{111\}$  planes can also produce second and third order twins and is observed within (110) and (111) epilayers [6].

Fig. 3 (a) shows the presence of (110) boundaries within (001) GaP epilayers. To verify that these are indeed inversion domain boundaries (IDBs) we have simulated the lattice image of a GaP crystal containing (110) type IDBs by translating the right side of the crystal by  $a/4\langle 111 \rangle$  with respect to the left [6]. This translation is performed to change the occupation of the respective fcc sublattices across the boundary. Thus a Ga atom would be placed on a P position after the translation leading to Ga-Ga and P-P wrong bonds at the IDB. The simulated image for a through thickness series obtained near Scherzer defocus are shown in Fig. 3 (b). These micrographs show that for a thin crystal  $< 4$  nm, the experimental lattice images do not show any evidence of the IDB. However, for thicknesses in excess of 4 nm, Scherzer images do indicate a discernible displacement across the boundary along a  $[001]$  direction, which gets accentuated with increasing thickness. Note that the black spots (which correspond to a doublet of Ga and P atoms) gets shifted downwards. The experimental lattice image in the insert of Fig. 3 (a) also reveals a slight displacement along the  $[001]$  direction for the black spots, consistent with our simulated images. The strain field associated with the formation of Ga-Ga and P-P bonds instead of Ga-P bonds produces rigid body displacements which give rise to the perceived contrast in lattice images.

GaP islands on (001) Si faceted on  $\{111\}$  planes take on the shape depicted in Fig. 4 (a). The P-terminated  $\{111\}$  facets have a higher density of dangling bonds compared to Ga-terminated  $\{111\}$  facets and this results in a higher surface energy and lower surface area. If the Si surface is uniformly covered by P atoms (i.e., the first atomic layer is P), then islands can coalesce on the same Si terrace as shown in Fig 4 (b) (i) and (ii). The schematic suggests that even if we were to assume wrong bonds are formed, these would not lie within a specified crystallographic plane or direction and would induce a high energy situation. The situation changes when islands coalesce across a surface step as indicated in Fig. 4 (b) (iii) and (iv). An IDB can form when a Ga-terminated facet coalesces either with another Ga-terminated facet across the step or a P-terminated facet across the step. In this case, by maintaining wrong bonds

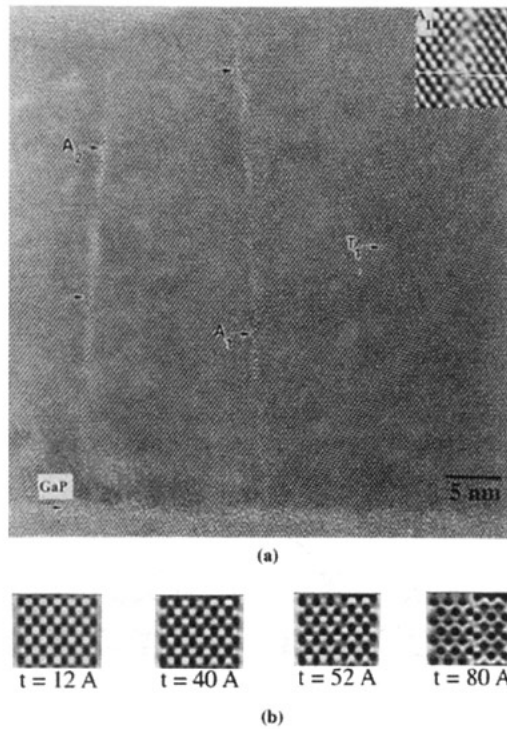
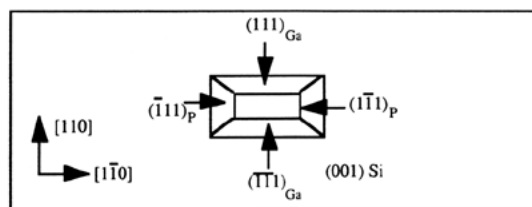


Fig. 3. (a) HRTEM image of columnar defects identified as IDBs in (001) GaP epilayers. The insert shows the displacement of black spots in the [001] direction. (b) Through thickness image simulations of an IDB in GaP imaged near Scherzer defocus.

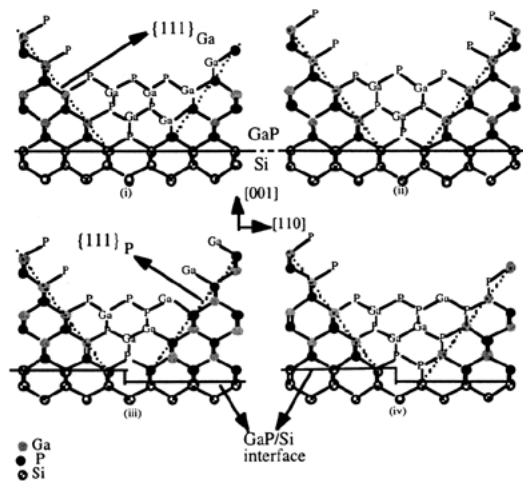
within the  $\{110\}$  plane perpendicular to the interface plane, one can maintain IDB free regions elsewhere. Thus, we can clearly see that a  $\{110\}$  type IDB will not form by island coalescence on the same terrace.

### CONCLUSIONS

The exposure of shallowly inclined P-terminated  $\{111\}$  facets whether on 2D or 3D islands is detrimental to the planar defect within epitaxial films. This explains the highly defective morphology within films grown on all four Si orientations. Amongst the four orientations,  $\{111\}$  epilayers are the most defective and this is attributed to the complex island shape and higher number of P-terminated facets within each island. Irregular growths are observed contiguous to continuously faulted segments and are formed due to multiple twinning on  $\{111\}$  facets. IDBs are observed in high resolution lattice images and are attributed to island coalescence across monatomic steps.



(a)



(b)

Fig. 4. (a) Schematic of a GaP island on (001) Si with  $\{111\}$  facets. (b) (i), (ii) depict island coalescence on the same terrace and (iii), (iv) depict island coalescence across a monatomic step.

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